UCRL-ID-113867-99

Environmental Protection Department

Operations and Regulatory Affairs Division

LLNL NESHAPs 1998 Annual Report

Lawrence Livermore National Laboratory
University of California Livermore, California 94550

LLNL NESHAPs
1998 Annual Report

Arthur H. Biermann
Gretchen M. Gallegos
Robert J. Harrach
Nicholas A. Bertoldo
Linda C. Hall
Robert L. Berger
Kris A. Surano

Contributors:

Frank J. Gouveia
Paula J. Tate
Paris E. Althouse
Judy L. Kelly

U.S. Department of Energy
Radionuclide Air Emission Annual Report
(under Subpart H of 40 CFR Part 61)
Calendar Year 1998

Site Name: Lawrence Livermore National Laboratory

Operations Office Information

Office: U.S. Department of Energy
Livermore Site Office

Address: 7000 East Avenue, L-293
Livermore, CA 94550

Contact: Steve Lasell Phone: 925-424-9784

Site Information

Operator: University of California

Address: 7000 East Avenue
Livermore, CA 94550

Contact: C. Susi Jackson Phone: 925-423-6577

June 1998

Table of Contents

Lawrence Livermore National Laboratory NESHAPs 1998 Annual Report

This annual report is prepared pursuant to the National Emissions Standards for Hazardous Air Pollutants (NESHAPs) 40 CFR Part 61, Subpart H; Subpart H governs radionuclide emissions to air from Department of Energy (DOE) facilities.

SYNOPSIS

NESHAPs limits the emission of radionuclides to the ambient air from DOE facilities to levels resulting in an annual effective dose equivalent (EDE) of 10 mrem (100 μSv) to any member of the public. The EDEs for the Lawrence Livermore National Laboratory (LLNL) site-wide maximally exposed members of the public from 1998 operations are summarized here.

- Livermore site: 0.055 mrem (0.55 μSv) (57% from point-source emissions, 43% from diffuse-source emissions). The point-source emissions include gaseous tritium modeled as tritiated water vapor as directed by EPA Region IX and is used for compliance purposes. LLNL believes a more realistic dose for the Livermore site is 0.049 mrem (0.49 μSv) (52% from point-source emissions, 48% from diffuse-source emissions). This dose is based on an assessment that represents a more realistic behavior of tritium gas in the environment (see discussion on page 10).

- Site 300: 0.024 mrem (0.24 μSv) (78% from point-source emissions, 22% from diffuse-source emissions).

The EDEs were generally calculated using the EPA-approved CAP88-PC air-dispersion / dose-assessment model. Site-specific meteorological data, stack flow data, and emissions estimates based on radionuclide inventory data or continuous-monitoring systems data were the specific input to CAP88-PC for each modeled source.

SECTION I. Facilities Information

Site Description

The University of California operates LLNL for DOE. LLNL was established in 1952 to conduct weapons research and development. LLNL's mission is to serve as a national resource in science and engineering, with a special responsibility for nuclear weapons. Laboratory activities focus on global security, energy, global ecology, biomedicine, economic competitiveness, and science and mathematics education. The Laboratory's mission is dynamic and has been broadened over the years to meet new national needs. LLNL consists of two sites—the main laboratory site located in Livermore, California (Livermore site), and the Experimental Test Facility (Site 300) located near Tracy, California. Figure 1 shows the locations of the sites.

Livermore site: LLNL's Livermore site occupies an area of 3.3 km^2 located about 60 km east of San Francisco, California, adjacent to the City of Livermore in the eastern part of Alameda County. More than 6 million people live within 80 km of LLNL; approximately 68,000 of them live in the City of Livermore.

The Livermore site is located in the southeastern portion of the Livermore Valley, a topographical and structural depression oriented east-west within the Diablo Range of the California Coast Range Province. The Livermore Valley forms an irregularly shaped lowland area approximately 26 km long and an average of 11 km wide. The floor of the valley slopes from an elevation of approximately 200 m at the eastern end to approximately 90 m at the southwest corner.

The climate of the Livermore Valley is characterized by mild, rainy winters and warm, dry summers. The mean annual temperature is about 15°C. Temperatures typically range from –5°C during some pre-dawn hours in the winter, to 40°C on a few summer afternoons. The 1998 annual wind data for the Livermore site are shown in Table 1 and displayed as a wind rose in Figure 2. Although winds are variable, the prevailing wind direction is from the southwest, especially during the summer. However, during the winter, the wind often blows from the northeast. Most precipitation occurs as rain between October and April with very little rainfall during the summer months. In 1998, the Livermore site received 522 mm of precipitation.

Site 300: Site 300, LLNL's Experimental Test Site, is located 24 km east of the Livermore site in the Altamont Hills of the Diablo Range and occupies an

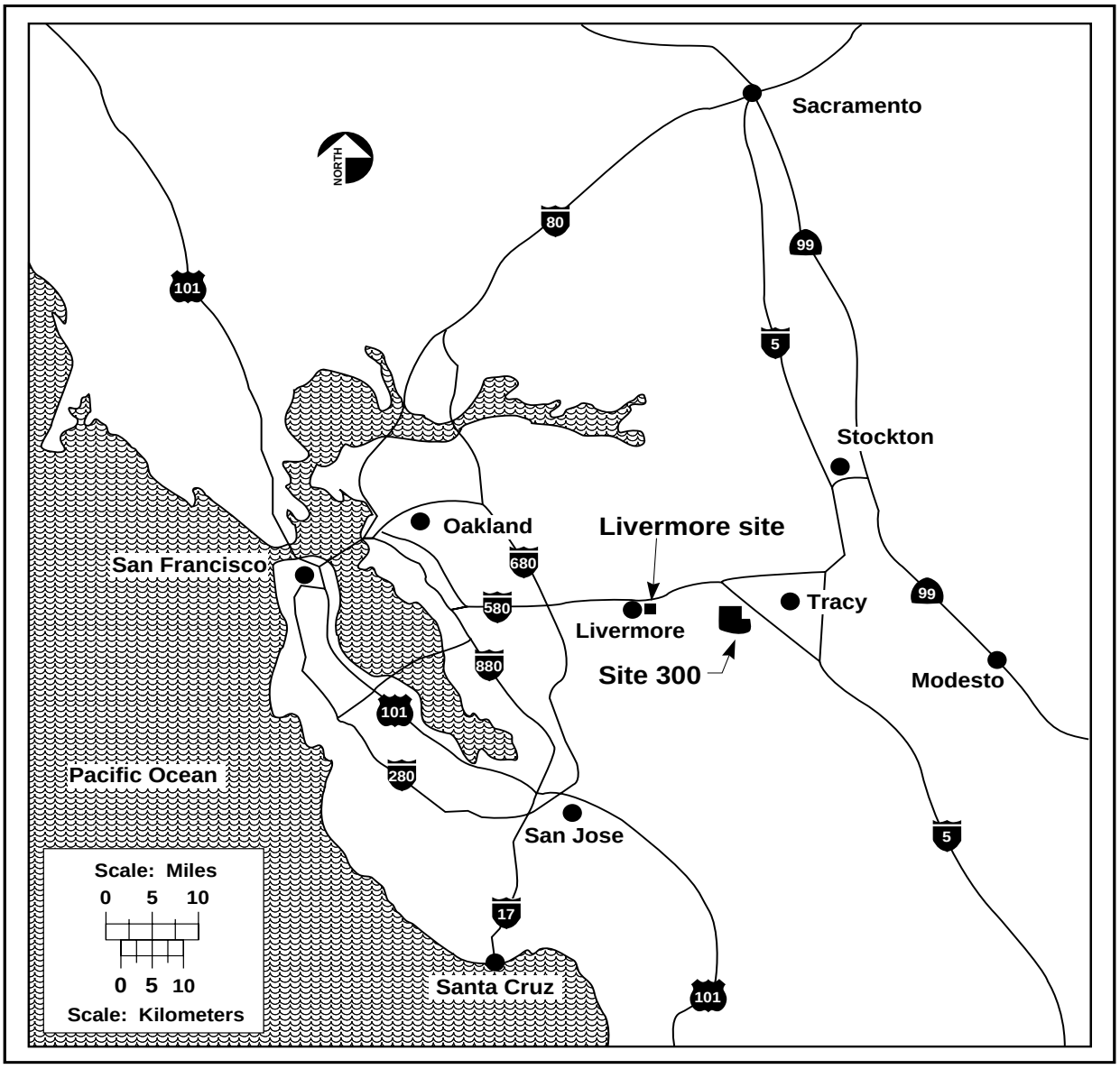

Figure 1. Locations of LLNL Livermore site and Site 300.

area of 30.3 km^2. It is close to two other explosives-testing facilities; one operated by Primex Physics International, the other by SRI International. A State of California vehicular-recreation area is located nearby, and wind-turbine generators line the surrounding hills. The remainder of the surrounding area is in agricultural use, primarily pasture land for cattle and sheep. The nearest residential area is the City of Tracy (population approximately 46,000), located 10 km to the northeast.

Table 1. Wind rose for LLNL's Livermore site at the 10-m level for 1998. Values are frequency of occurrence (in percent). Columns and rows may not exactly sum to the listed totals due to rounding.

	Wind Speed Range (m/s)					
Direction	0.0-0.40	0.50-2.90	3.00-4.90	5.00-6.90	=>7.00	Total
NNE	0.24	2.89	1.27	0.03	0.00	4.4
NE	0.24	3.92	1.55	0.05	0.00	5.8
ENE	0.24	3.49	0.07	0.00	0.00	3.8
E	0.24	3.55	0.00	0.00	0.00	3.8
ESE	0.24	3.27	0.03	0.00	0.00	3.5
SE	0.24	2.78	0.13	0.06	0.00	3.2
SSE	0.24	2.32	0.31	0.07	0.00	2.9
S	0.24	5.72	0.93	0.30	0.12	7.3
SSW	0.24	6.73	2.75	0.70	0.15	10.6
SW	0.24	6.51	8.27	2.73	0.23	18.0
WSW	0.24	6.97	5.21	1.55	0.03	14.0
W	0.24	5.99	5.38	0.77	0.00	12.4
WNW	0.24	2.24	1.01	0.09	0.00	3.6
NW	0.24	1.88	0.17	0.00	0.00	2.3
NNW	0.24	1.85	0.23	0.06	0.02	2.4
N	0.24	1.22	0.31	0.12	0.05	1.9
Total	3.9	61.3	27.6	6.5	0.6	100.0

Table 2. Wind rose for LLNL's Site 300 at the 10-m level for 1998. Values are frequency of occurrence (in percent). Columns and rows may not exactly sum to the listed totals due to rounding.

	Wind Speed Range (m/s)					
Direction	0.0-0.4	0.5-4.9	5.0-6.9	7.0-10.9	≥11.0	Total
NNE	0.09	1.55	0.15	0.03	0.07	1.9
NE	0.09	2.10	0.31	0.00	0.00	2.5
ENE	0.09	1.99	0.51	0.08	0.00	2.7
E	0.09	1.87	0.25	0.02	0.00	2.2
ESE	0.09	2.41	0.15	0.08	0.00	2.7
SE	0.09	3.10	0.48	0.49	0.06	4.2
SSE	0.09	3.11	0.26	0.23	0.15	3.8
S	0.09	4.68	0.64	0.22	0.06	5.7
SSW	0.09	2.98	0.18	0.11	0.07	3.4
SW	0.09	2.89	1.11	3.11	1.36	8.6
WSW	0.09	4.01	6.86	13.35	1.93	26.2
W	0.09	4.80	4.48	2.52	0.05	11.9
WNW	0.09	3.07	1.28	0.11	0.00	4.6
NW	0.09	4.24	1.21	0.89	0.03	6.5
NNW	0.09	5.57	1.94	1.72	0.27	9.6
N	0.09	2.11	0.78	0.43	0.07	3.5
Total	1.4	50.5	20.6	23.4	4.1	100.0

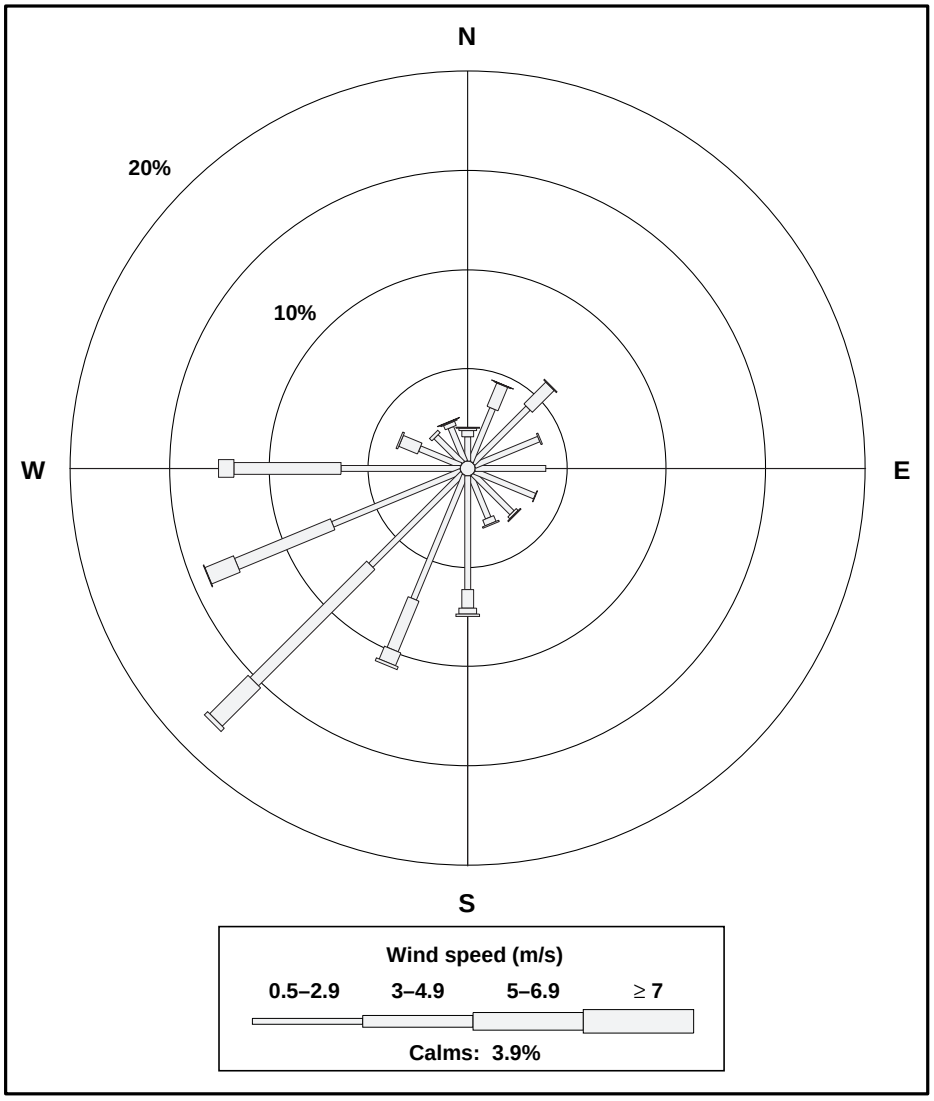

Figure 2. Wind rose showing the average annual wind speed, frequency of occurrence, and direction at the Livermore site, 1998.

The topography of Site 300 is much more irregular than that of the Livermore site; it consists of a series of steep hills and ridges, which are oriented along a generally northwest/southeast trend, separated by intervening ravines. The elevation ranges from approximately 540 m in the northwestern portion of the site to 150 m at the southeast corner. The climate at Site 300 is similar to that of the Livermore site, with mild winters and dry summers. The complex topography of the site significantly influences local wind and temperature patterns, making the temperature range somewhat more extreme than at the Livermore site. The 1998 annual wind data for Site 300 are shown in Table 2 and

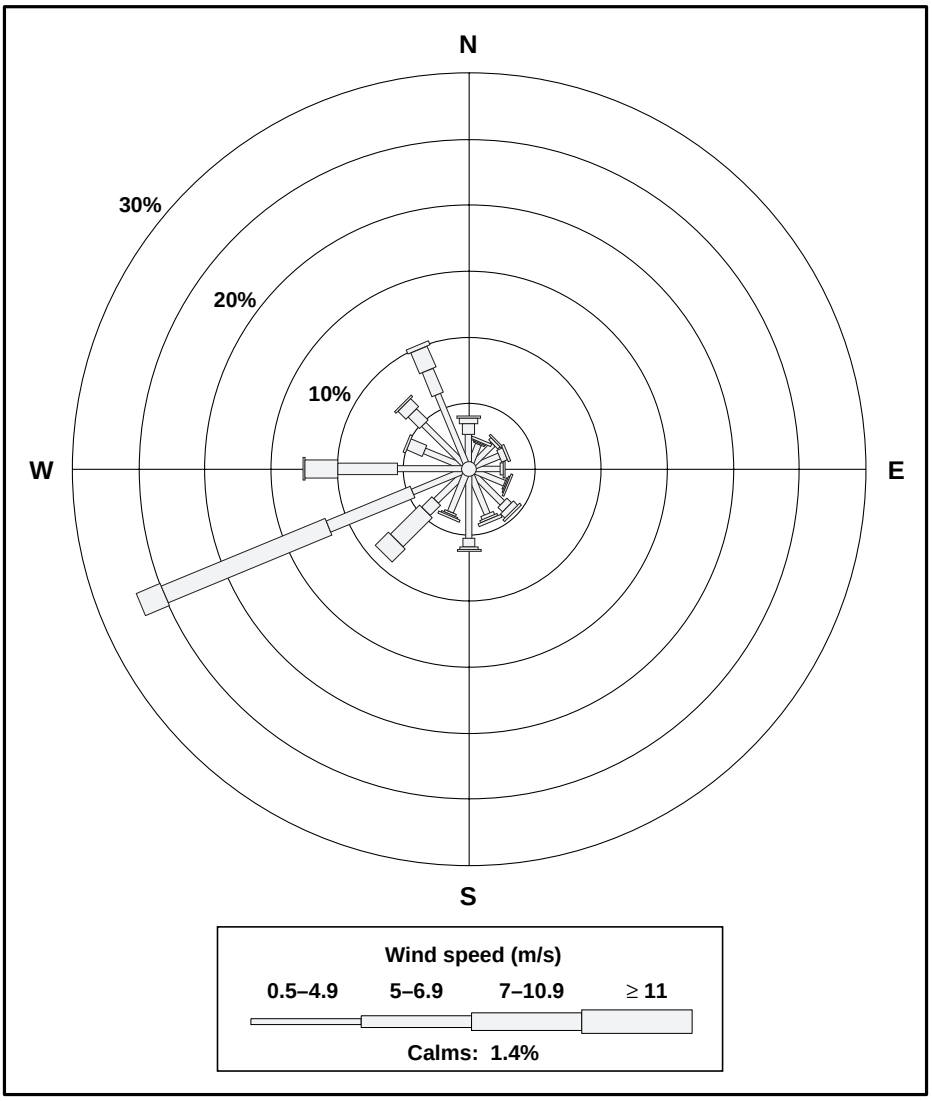

Figure 3. Wind rose showing the average annual wind speed, frequency of occurrence, and direction at Site 300, 1998.

displayed as a wind rose in Figure 3. Prevailing winds are from the west-southwest. As is the case at the Livermore site, precipitation is highly seasonal, with most precipitation occurring between October and April. Site 300 received 475 mm of precipitation during 1998.

Source Description

Many different radioisotopes are used at LLNL for research purposes, including transuranics, biomedical tracers, tritium, mixed fission products, and others (Table 3). Radioisotope-handling procedures and work enclosures are determined for each project, depending on the isotopes, the quantities being used, and the types of operations being performed. Radioisotope handling and working environments include glove boxes, exhaust hoods, and laboratory bench tops. Exhaust paths to the atmosphere range from triple-HEPA (High-Efficiency-Particulate-Air)-filtered stacks, to roof vents and stacks lacking abatement devices, to direct dispersal of depleted uranium during explosives testing at Site 300, to a variety of diffuse-area sources.

Table 3. Radionuclides used at LLNL during 1998.

^{3}H	^{60}Co	^{133}Ba	^{209}Po	^{234}Th	^{241}Am
^{13}N	^{63}Ni	^{134}Cs	^{226}Ra	^{234}U	^{241}Pu
^{14}C	^{88}Y	^{137}Cs	^{228}Ra	^{235}U	^{242}Pu
^{15}O	^{90}Sr	^{144}Ce	^{228}Th	^{236}Pu	^{243}Am
^{22}Na	^{90}Y	^{147}Pm	^{229}Th	^{236}U	^{244}Cm
^{32}P	^{95}Nb	^{149}Eu	^{230}Th	^{237}Np	^{249}Cf
^{33}P	^{99}Tc	^{151}Sm	^{231}Pa	^{238}Pu	^{252}Cf
^{35}S	106Ru	^{152}Eu	^{231}Th	^{238}U	
^{54}Mn	^{125}I	^{154}Eu	^{232}Th	^{239}Np	
^{57}Co	^{125}Sb	^{155}Eu	^{232}U	^{239}Pu	
^{59}Ni	^{131}I	^{207}Bi	^{233}U	^{240}Pu	

SECTION II. Air-Emission Data

Sources

At LLNL, areas where radioactive materials are used or stored, or where activation products occur, are called Radioactive Materials Management Areas (RMMAs). Detailed information is given in Attachment 1 for point-source emissions from the Livermore-site RMMAs in which radiological operations took place during 1998. Building 514 and five other Livermore-site sources external to buildings (including the RMMA at the Building 612 Hazardous Waste Management Yard) are treated as diffuse-area sources.

Similarly, detailed information is given in Attachment 1 for experiments at two Site 300 explosives-testing facilities (Buildings 801 and 851 and their associated firing tables). Site 300 is also treated as a diffuse-area source of residual tritium and depleted uranium contamination.

1998 Inventory Update and Effective Dose Equivalent (EDE) Calculations

For this year's report, covering activities in 1998, we updated the radionuclide inventories in key facilities, defined as those that accounted for 90% of the 1997 Livermore site radiological dose to members of the public. We also inventoried all RMMAs that began operations in 1998. Radionuclide inventory forms, with guidance for completing them, were sent to the unmonitored facilities that contributed to 90% of the dose in 1997 and to new unmonitored facilities having the potential for radionuclide emissions to the air. The forms were completed by experimenters, and certified by facility managers. Radionuclide inventories for all Site 300 explosives experiments and assessments of source terms for known diffuse sources at both sites were also updated.

Dose-assessment modeling runs were conducted for all diffuse sources and for all point sources using measured radionuclide releases to air, or potential releases based on radionuclide inventory data. The model used was CAP88-PC (see Section III); we incorporated 1998 on-site meteorological data (wind, precipitation, and temperature) along with the 1998 radionuclide inventory or monitoring data. Annual dose is reported as whole-body EDE expressed in units of mrem (followed by μSv). When reasonable to do so, modeling runs were combined by building, rather than a separate model run for each stack or room. This is permitted by the 1995 Memorandum of Understanding between the U.S. EPA and the DOE concerning radionuclide NESHAPs.

A generalized description of each facility and its operations is provided in Attachment 1. The following information is shown for each listed emission point or stack:

- Building and room number(s)
- Specific stack identification code(s)
- Generalized operations in the room(s) or area(s)
- Radionuclides utilized during 1998
- Annual radionuclide inventory with potential for release (by isotope, in curies)
- Physical-state factors (by isotope)
- Stack parameters
- Emission-control devices and emission-control-device abatement factors
- Estimated or measured annual emissions (by isotope)
- Distance and direction to the site-wide maximally exposed individual (SW-MEI)
- Calculated EDE to the SW-MEI
- Distance and direction to the maximally exposed individual for that specific source (MEI)
- Calculated EDE to the MEI (source term not adjusted for emission controls)
- Source category

A more complete description of these terms is provided in the introductory material to the attachment.

The radionuclides shown in the attachment are those from specific emission points where there was a potential for air emissions. If radionuclides were present, but encapsulated or sealed for the entire year, radionuclides, annual inventories, and emissions are not listed.

Actual measurements of air radioactivity and effluent flow are the basis for reported emissions from continuously monitored sources. LLNL facilities that had continuously monitored discharge points are Buildings 175, 177, 251, 292, 331, 332, 490, and 491. Discharge points at Buildings 175, 177, 251, 292, 332, 490, and 491 are monitored for gross alpha and gross beta activity. Building 331 discharges are monitored for tritium.

Operations in the Tritium Facility (Building 331) released a total of 4.1×10^{12} Bq (110 Ci) of tritium. Of this, approximately 3.1×10^{12} Bq (85 Ci) were released as tritiated water (HTO). The remaining 18% of the tritium released, 9.3×10^{11}

Bq (25 Ci), was elemental tritium gas (HT). The highest single weekly stack emission from the facility was 3.2×10^{11} Bq (8.7 Ci), of which 1.5×10^{11} Bq (1.6 Ci) was HTO.

Modeling of the HTO emissions from the Tritium Facility using CAP88-PC results in an estimated dose to the SW-MEI of 0.023 mrem (0.23 µSv). However, modeling of the dose caused by HT emissions is more complicated, requiring knowledge of the oxidation of HT to HTO in the environment. Oxidation is required for the HT emissions to be of consequence since the accepted dose conversion factor for HT is 25,000 times lower than that of HTO. Modeling of the oxidation process and dose from HT emissions is not described by CAP88-PC.

Recently, as directed by U.S. EPA Region IX, we have evaluated the combined HT and HTO emissions from the Tritium Facility in 1998 as if they were all HTO. The resulting dose to the SW-MEI from the combined emissions is 0.029 mrem (0.29 µSv). We believe and EPA Region IX acknowledges that this dose, based on compliance rules, is a very conservative overestimate of the actual dose and not indicative of physical reality.

Our preliminary research into the oxidation of HT to HTO in the environment has shown the oxidation mostly occurs slowly in the soil after significant dispersion followed by deposition of the lighter-than-air HT gas (Brown, Ogram and Spencer, 58 Health Physics, 171-181(1990)). The contribution of oxidation of tritium gas to tritium water in the atmosphere is small. Only tritium gas in the T_2 form is oxidized (Noguchi, 27 Transactions of Fusion Technology, 56-61 (1995)). The residence time of tritium in the atmosphere has been estimated at 6.5 years (Momoshima, Okai, Kajiand Takashima, 54 Radiochemica Acta, 129-132 (1991)). In addition, we have found no scientific literature that suggests a rapid conversion of tritium gas to tritiated water vapor in the environment. Therefore, we believe the approach of estimating dose by assuming all tritium emitted is HTO significantly overestimates the SW-MEI dose. While this approach correctly errs on the conservative side by overestimating the dose, we believe that more work is warranted to truly understand the HT transition process and then to develop a more accurate dose estimating tool.

We are currently involved in discussions with the U.S. EPA, the DOE, and the international health physics community about the appropriateness of modeling HT as HTO. We intend to report on the results of our continued investigation in our next NESHAPs annual report.

For most of the discharge points from the other facilities, results from continuous sampling are below the minimum detectable concentration (MDC) of the analysis; sometimes as few as 1 to 4 samples (out of 25 to 50 per year) have concentrations greater than the MDC. Generally, these few samples having results above the MDC are only marginally above the MDC. Use of zero values for this type of data can be justified based on knowledge of the facility, the use of multiple-stage HEPA filters in all significant release pathways, and alpha-spectroscopy-based isotopic analyses of selected air-sampling filters. These isotopic analyses demonstrate that detected activity on air-sampling filters comes from naturally occurring radionuclides, such as radon daughters, e.g., polonium, on the air sampling filters. In addition, because of exhaust configurations at some facilities, the monitoring systems sometimes sample air from the ambient atmosphere along with the HEPA-filtered air from facility operations giving rise to background atmospheric radioactivity being collected. Because of these considerations, the emissions from such facility operations are reported as zero. Furthermore, even if the MDC values are used in calculations of the emission estimates for these facilities, which would be an extremely conservative approach, the total dose attributable to LLNL activities is not significantly affected.

In 1998, samples from four emission points at Building 251 (the unhardened area) yielded gross alpha results greater than the MDC on a significant number of the samples collected throughout the year. We use gross alpha as the primary indicator of potential emissions for operations such as those at Building 251 that involve the use of uranium and transuranic materials. Gross beta results are used as a further corroboration of those gross alpha results having concentrations above the MDC. The gross alpha monitoring concentrations for Building 251 ranged from -3.0×10^{-4} Bq / m^3 (-8.2×10^{-15} Ci / m$^{3)}$ to 1.4×10^{-3} Bq / m^3 (3.7×10^{-14} Ci / m^3). Because of the number of samples with values above the MDC, we have taken a conservative approach and are reporting gross alpha and gross beta measurements as actual emissions. The gross alpha and gross beta emissions for Building 251 were determined to be 2.7×10^3 Bq / y (7.3×10^{-8} Ci / y) and 2.3×10^4 Bq / y (6.1×10^{-7} Ci / y. The resulting radiological dose due to the reported values is 1.3×10^{-4} mrem (1.3×10^{-3} μSv), less than the dose due to other facility emissions at the Livermore site.

SECTION III. Dose Assessment

Description of Dose Model

Estimates of individual and collective radiological doses to the public from all point sources and most diffuse sources at LLNL were obtained using the EPA-developed computer code CAP88-PC. The four principal pathways—internal exposures from inhalation of air, ingestion of foodstuffs and drinking water, external exposures through irradiation from contaminated ground, and immersion in contaminated air—are evaluated by CAP88-PC. The doses are expressed as whole-body effective dose equivalents (EDEs), in units of mrem/y (1 mrem = 10 μSv). Separate doses for the Livermore site and Site 300 from point-source emissions (i.e., stack emissions) and diffuse-source emissions at the two sites are reported.

Three potential doses are emphasized: (1) The dose to the site-wide maximally exposed individual (SW-MEI), which combines the effects of all emission points, for comparison to the 10 mrem/y (100 μSv/y) standard; (2) the maximum dose to any member of the public (assumed to be at the LLNL fence line), in any direction, due to each unabated emission point on the site to determine the need for continuous monitoring; and (3) the collective dose to the populations residing within 80 km of the two LLNL sites, adding the products of individual doses received times the number of people receiving them.

Summary of Model Input Parameters

General Model Inputs: Attachment 1 details the key identifiers and input parameters for the CAP88-PC model runs. These include building number; stack ID; isotope(s); emission rate in curies per year (1 Ci = 3.7×10^{10} Bq); and stack parameters, including height, diameter, and emission velocity.

Meteorological Data: All model runs used actual 1998 Livermore-site and Site 300 meteorological data, collected from the meteorological towers for each site. At these towers, wind speed and direction are sampled every few seconds, temperature every minute; and all are averaged into quarter-hour increments, time-tagged, and computer-recorded. The data are converted into a CAP88-PC input wind file using EPA guidelines.

Surrogate Radionuclides: CAP88-PC contains a library of 265 radionuclides; however, it does not contain all the radionuclides in use at LLNL. As a consequence, it was necessary in a few cases to use surrogate radionuclides to

estimate EDEs. Attachment 2 shows the surrogate radionuclides used in CAP88-PC. In selecting the surrogates, the most-restrictive lung class (whether clearance from the lungs takes place in days, weeks, or years) was used. When possible, a surrogate radionuclide with similar chemistry and similar values for "annual limits of intake via inhalation and derived air concentration," as specified in the EPA's Federal Guidance Report No. 11 was used. In some cases, experimenters did not have isotopic analyses of mixtures of radionuclides and could only identify their radionuclide inventory as "gross alpha," "gross beta," "gross gamma," or "mixed fission products" (MFP). In these cases, ^{239}Pu was used as the surrogate for gross alpha, ^{137}Cs was used as the surrogate for gross gamma, and ^{90}Sr was used as the surrogate for gross beta and mixed fission products to provide conservative dose estimates.

Population Inputs: Population distributions centered on the two LLNL sites were compiled from 1990 census data. The population data files (distribution of population with distance and direction) used in the 1998 modeling effort are described in Section VI under "Collective Effective Dose Equivalent."

Land-Use and Agricultural Inputs: Options for model inputs regarding agricultural characteristics and land use are established by the EPA, and the particular designation selected can strongly influence the ingestion dose received by the population being evaluated. Following our investigation in 1995 into the use of the various options, the "user entered" option was again selected for the CAP88-PC modeling effort for 1998. The values entered corresponded to the "local agriculture" option (i.e., everything is home produced), with one exception—all milk consumed was assumed to be imported for individual dose assessment. The assumption that all milk comes from local cows is not supported by the agricultural activities conducted in the area. A detailed discussion of how the dose from tritium is calculated by CAP88-PC is presented in the LLNL NESHAPs 1995 Annual report (Gallegos et al., 1996, Lawrence Livermore National Laboratory, UCRL-ID-113867-96).

Emission Source Terms: The source term(s) from each emission point in the calculations was determined by one of two methods: For continuously monitored sources, the sampling data (curies released per unit time) for each radionuclide were used directly. For unmonitored facilities, the radionuclide inventories, together with time factors and EPA-specified physical state factors, are used to estimate the potential emissions to air from a source. The time factors are used to adjust for the fact that the radionuclide may not always be in the same facility all year or may be encapsulated or enclosed for a substantial part of the year. The time factors are chosen to allow a reasonable

estimate of the amount of radioactive material that may potentially be released into the atmosphere. The EPA-specified factors for potential release to air of materials in different physical states (solid, liquid, powder, or gas) are those stated in 40 CFR Part 61, Appendix D. If the material was an unconfined gas, then the fraction 1.0 was used; for liquids and powders, 1.0×10^{-3} was used; and for solids, 1.0×10^{-6} was used. In 1996, U.S. EPA granted approval for LLNL to use alternative emission factors for elemental uranium as follows: an emission factor of 1×10^{-6} can be used for elemental uranium heated at temperatures below 1100°C, an emission factor of 1×10^{-3} can be used for elemental uranium heated at temperatures below 3000°C, and an emission factor of 1 shall be used for temperatures greater than 3000°C. These factors are allowed provided that the uranium is not intentionally dispersed to the environment and that the processes do not alter the chemical form of the uranium. The physical-state-dependent release fraction and the time factor are used to adjust (by multiplication) the total annual inventory to yield the potential annual release to air. In addition, emission-control abatement factors (40 CFR 61, Appendix D), when applicable, were applied. Each HEPA filter stage was given a 0.01 factor. (However, abatement factors were not used to evaluate compliance with the 0.1 mrem standard that determines the need for continuous monitoring at a facility.) The use of actual monitoring data is much more direct, and presumably more accurate, than using assumptions based on inventory, time factors, release fractions, and emission-control factors.

Site-Wide Maximally Exposed Individual: For LLNL to comply with the NESHAPs regulations, the LLNL site-wide maximally exposed individual cannot receive an EDE greater than 10 mrem/y (100 μSv/y). The site-wide maximally exposed individual (SW-MEI) is defined as the *hypothetical* member of the public at a single residence, school, business, or office who receives the greatest LLNL-induced EDE from the combination of all radionuclide source emissions.

To determine the location of the 1998 SW-MEI, CAP88-PC results from multiple sources were combined. Sources were selected to include those expected to give significant contributions to the EDE. These consisted of Building 331 point and diffuse sources and the Building 612 diffuse source. Because EDE results from CAP88-PC are relative to the location of the specified source, direct summing of results from multiple sources can only be accomplished using an interpolation method. To do this, the location of each selected source relative to a common location (the Livermore-site center) and a set of receptor locations (where the combined EDEs from the selected

sources were to be evaluated), also relative to the site center, were specified in the modeling efforts that supported determination of the SW-MEI. The receptor locations included 48 equally spaced directions from the site center and 4 additional receptor locations along the eastern Livermore-site boundary. The interpolation method was used to calculate the EDEs for the desired set of receptor locations for each source. These resulting interpolated EDEs for each source, now for the same set of locations, were then summed, and the SW-MEI determined.

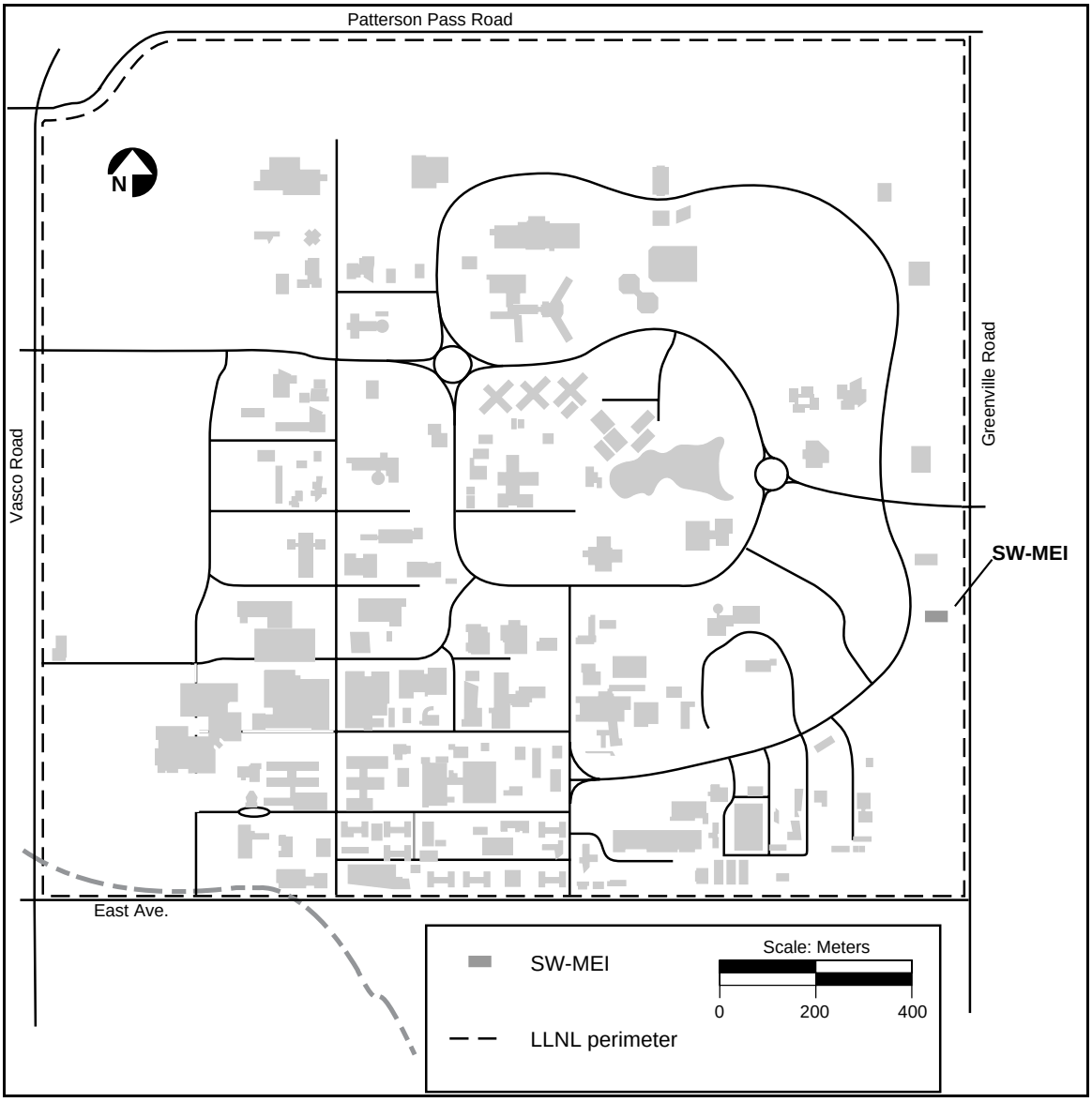

Figure 4. Location of Site-Wide Maximally Exposed Individual (SW-MEI) at the Livermore site, 1998.

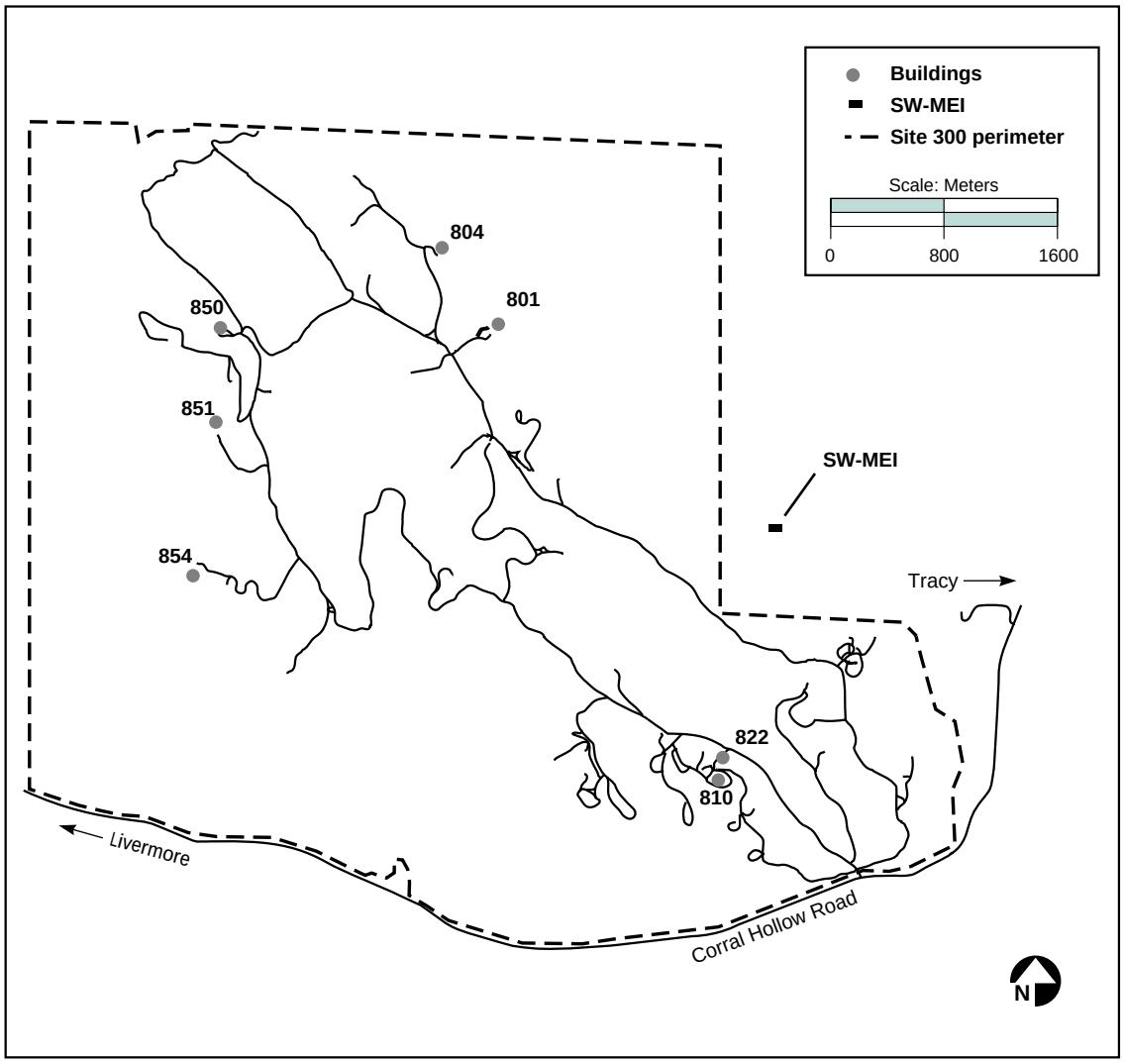

Figure 5. Location of Site-Wide Maximally Exposed Individual (SW-MEI) at Site 300, 1998.

At the Livermore site, the SW-MEI for 1998 was located at the UNCLE Credit Union, about 10 m outside the controlled eastern perimeter of the site, as shown in Figure 4.

At Site 300, the 1998 SW-MEI was located in an experimental area termed "Bunker 2" operated by Primex Physics International. Bunker 2 lies about 300 m outside the east-central boundary of Site 300, as shown in Figure 5. This bunker is approximately 2.4 km east southeast of the firing table at Building 801.

In Attachment 1, the distance and direction to the respective SW-MEI are shown for each facility at each site. Doses to the site-specific SW-MEIs were evaluated for each source and then totaled for site-specific evaluations against the 10 mrem/y dose standard (see "Total Dose Estimate" in Section IV).

Maximally Exposed Public Individual: To assess compliance with the requirement for continuous monitoring (potential dose greater than 0.1 mrem/y {1.0 μSv/y}), emissions must be individually evaluated from each point source; the location of the maximally exposed public individual (MEI) is generally different for each emission point. The maximum dose at a location of unrestricted public access typically occurs at a point on the site perimeter. Therefore, it is often referred to as the maximum "fence line" dose, although the off-site maximum dose could occur some distance beyond the perimeter. (This could happen, e.g., when the perimeter is close to a stack; however, for all emission points at the Livermore site and Site 300, calculations show that ground-level concentrations of radionuclides decline monotonically beyond LLNL boundaries.) As stipulated by the regulations in 40 CFR Section 61.93 (b)(4)(ii), modeling for assessment of continuous monitoring requirements assumed unabated emissions (i.e., no credit was taken for emission abatement devices, such as filters). Attachment 1 provides, for each point source, the dose to the MEI and the distance and direction to the LLNL fence line where the MEI is located.

Special Modeling Challenges: Among the sources at LLNL, explosives tests using depleted uranium at Site 300 and diffuse sources at both sites required special attention.

Site 300 Explosives Experiments: During Site 300 explosives experiments, some of the explosives assemblies contain depleted uranium. The explosives assemblies are placed on an open-air firing table and detonated. Only limited data are available to characterize the initial state of the cloud of explosive decomposition products created by the detonation because properties of the cloud are not routinely measured in the experiments. Empirical scaling laws can be used, however, to define the cloud using the radionuclide and explosives inventories. Isotopic ratios for depleted uranium are used; the three uranium isotopes with atomic weights 238, 235, and 234 occur in the weight percentages 99.8, 0.2, and 5×10^{-4}. Their masses are multiplied by their specific activities to determine the total activity for each isotope in the cloud. It is assumed that all the uranium is dispersed into the cloud, and the median particle size is assumed to be the CAP88-PC default value of 1 μm. The assumption that all uranium is aerosolized and dispersed as a cloud results in a highly conservative off-site dose estimation—we

believe a more realistic release-to-air fraction for the uranium is no greater than 0.2, but we lack sufficient data to use a value other than 1.0. CAP88-PC simulates each shot as a low-level, steady-state, stack-type emission occurring over one year. An alternative modeling methodology for treating these short-duration explosive events was submitted for approval in 1992, but LLNL was directed by EPA to use the CAP88-PC code for these calculations.

Diffuse Sources: Diffuse emissions are generally area sources external to buildings, as discussed in Section IV, below. The dose assessments for diffuse sources can be derived from modeling based on radionuclide-inventory data, or can be determined from environmental-surveillance monitoring data.

Modeling Documentation: Copies of individual model runs, including input parameters and resultant calculated doses, are on file with the Terrestrial & Atmospheric Monitoring & Modeling Group (TAMM) of the Environmental Protection Department at LLNL.

Point Source Summary

The 1998 calculated EDE to the SW-MEI from Livermore-site point sources was 0.031 mrem (0.31 µSv). The dose from point sources includes HT emissions modeled as HTO as directed by EPA Region IX. EPA Region IX acknowledges that such modeling results is a very conservative overestimation of the dose. This methodology is used for purposes of compliance and we do not believe it provides a technically accurate dose estimate. In any case, the 1998 dose is lower than the 1997 reported EDE from Livermore-site point sources of 0.078 mrem (0.78 µSv). The reduction in EDE to the SW-MEI can be attributed to reduced emissions from the Tritium Facility (Building 331) where emissions accounted for 0.075 mrem (0.75 µSv) in 1997 but only 0.029 mrem (0.29 µSv) in 1998 (again with HT emission modeled as HTO in 1998 only).

The calculated EDE to the SW-MEI at Site 300 was calculated to be 0.019 mrem (0.19 µSv) from point-source emissions. Nearly all of this dose resulted from Building 801 and Building 851 firing-table emissions in the course of explosives experiments. The 1998 EDE is an increase from the 0.011 mrem (0.11 µSv) dose modeled for 1997. The increase in dose is primarily the result of an increase in the quantity of depleted uranium used in the experiments. In October 1998, LLNL revised its estimate of dose from Site 300 operations in calendar year 1997. The revision was prompted by identification of a

miscommunication about the location of releases modeled at the site. Further discussion is provided in Section IV and Attachment 3.

All the dose evaluations from point-source emissions, and those from most diffuse sources discussed below, were made using the EPA-mandated CAP88-PC dispersion model. They result in levels of public exposure well below the EPA standard, which limits the whole-body EDE to members of the public from DOE activities to 10 mrem/y (100 μSv/y). Discussion of the contribution to EDE to members of the public from diffuse sources is presented in Section IV.

SECTION IV. Additional Information

Construction and Modifications

Proposed facilities and significantly modified operations are assessed for NESHAPs requirements during the National Environmental Policy Act (NEPA) process. Under NEPA, all proposed projects or actions that might involve NESHAPs issues or concerns—not just pertaining to radionuclides but to toxic air contaminants as well—are reviewed and evaluated. If the proposal includes operations that require a NESHAPs assessment, necessary modeling is conducted. If insufficient information is available for modeling at the time the NEPA documents are prepared, LLNL includes in the NEPA documents a statement that NESHAPs review, modeling, and monitoring requirements will be met. It is the responsibility of the individual project proponent to supply the specific information required for any NESHAPs modeling, analysis, and review that must be completed before operations described in the document are initiated.

In 1998, the Expedited Technology of Molten Salt Oxidation project, a mixed-waste segregation treatment study, began operations in Building 292. The project is designed to recover many different low-level radionuclides in the form of solid salts. The NESHAPs review of proposed operations indicated that continuous sampling of the air effluent was required (40 CFR 61.93), but that approval to construct or modify was not required (40 CFR 61.96). All exhausts from the project are routed through a single stack. A continuous filter-type sampler with a multi-nozzle probe was installed in the exhaust downstream of HEPA filters. Membrane filters, which are used to collect particulate emissions, are analyzed for gross alpha and gross beta activity.

Three new facilities are currently under construction. All of these facilities were assessed prior to construction for compliance with NESHAPs, and effluent sampling systems are planned for all three. These facilities are the Contained Firing Facility (CFF) at Site 300, and the National Ignition Facility (NIF) and the Decontamination Waste Treatment Facility (DWTF) at the Livermore site. The CFF project will allow containment of some explosives tests currently conducted outdoors at Site 300's Building 801. The CFF project consists of an enclosed firing chamber, a support facility and a diagnostic equipment facility. Phase II construction began in April 1999. The DWTF is a facility that will allow waste handling to occur in a facility that has improved air emissions control and will enable the handling of additional waste streams. Phase I construction (site preparation and installation of underground utilities) has been completed. Construction of the solid waste

processing building, the storage building, and the office building were completed in 1998. Construction of the building housing the stack and air handling systems and liquid waste processing building has not started due to permitting delays. The National Ignition Facility (NIF) will contain the world's largest laser, a research tool allowing scientists to recreate on earth conditions equivalent to the center of the sun. The NIF will focus 192 extremely powerful laser beams onto a BB-sized capsule of deuterium and tritium, forcing the two heavy isotopes of hydrogen to combine through compression and heating, producing ignition and self-sustained fusion burn. The NIF construction project began in 1996 and will end in 2003 with initial operations beginning in 2001. It is being designed, built and operated by a team from Lawrence Livermore, Los Alamos and Sandia National Laboratories and the University of Rochester. NIF construction is well underway. Major work has progressed on the laser building, laser and target bays, and the diagnostics and optics assembly buildings.

Unplanned Releases

There were no unplanned atmospheric releases of radionuclides at the Livermore site or Site 300 in 1998.

Diffuse Source Dose Assessments

Diffuse, or non-point, sources are difficult to quantify. There are no EPA-mandated methods for estimation or measurement, although LLNL did review a second draft of EPA guidance on this topic during 1994. At this time, however, dose calculations associated with this type of source remain left to the discretion of the DOE facility. Livermore-site and Site 300 diffuse sources are described separately.

Livermore-Site Diffuse Sources

The dose calculations from 1998 diffuse sources at the Livermore site required three different modeling approaches. Building 331 Yard and Building 612 Yard needed facility personnel knowledge and environmental-surveillance data to estimate emissions; Building 292 required vegetation monitoring and CAP88-PC modeling techniques; Building 514 required radiological-inventory data and CAP88-PC modeling techniques; and data from ambient-air monitoring were used to calculate the dose for the Southeast Quadrant.

Building 292: Elevated tritium concentrations in soil moisture near Building 292 resulted from a historic leak in an underground retention tank. This

contamination has resulted in diffuse tritium emissions due to transpiration from vegetation. In 1998, quarterly samples of the pine tree, which had previously been identified as the primary source of transpired tritium, were used to estimate the emission of tritium from this source. The maximum concentration of tritium in the tissue water of the pine tree was 2970 pCi/L (110 Bq/L) in 1998. Assuming the tree has an area of 79 m^2 and a transpiration rate of 190 L/d, the resulting emission rate from this source is 2.0×10^{-4} Ci/y (7.4×10^6 Bq/y). This estimated emission compares well with previous estimates, which ranged from 4.8×10^{-4} Ci/y (1.8×10^7 Bq/y) to 1.4×10^{-3} Ci/y (5.1×10^7 Bq/y) in 1994 through 1997. The current source term produced a calculated 1998 dose to the SW-MEI from the Building 292 area of 2.9×10^{-8} mrem (2.9×10^{-7} µSv).

Building 331 Yard: As the Tritium Facility (Building 331) conducts operations, tritium-contaminated equipment and material slated for disposal is removed from the building, packaged in a waste-accumulation area, and sent to Hazardous Waste Management Division (HWM) facilities. During 1998, outgassing from such waste processing released approximately 6 Ci (2.2×10^{11} Bq) of tritium to the atmosphere outside Building 331. The estimated releases were derived from measurements of surface contamination on the material, process and facility knowledge, and environmental-surveillance measurements. The estimated release was modeled in CAP88-PC as a 1 m^2 area source, leading to a calculated 1998 dose to the SW-MEI of 3.9×10^{-3} mrem (3.9×10^{-2} µSv).

Building 514: Another potential source of diffuse emissions of a variety of radionuclides was HWM waste-storage and treatment operations. Building 514 houses the HWM "tank farm," consisting of six 7,170-liter tanks with ancillary equipment such as pumps, mixers, probes, and a bulking station. The tanks are used to store and treat liquid and solid radioactive and/or mixed wastes. Treatment is performed on a batch basis. Chemicals and waste are added to the tanks to achieve the desired treatment objectives. A 1998 radionuclide inventory was conducted for the facility to determine the diffuse source term (Attachment 1). CAP88-PC modeling gave a 1998 EDE for the Tank Farm to the SW-MEI of 1.3×10^{-4} mrem (1.3×10^{-3} µSv).

Building 612 Yard: The Building 612 Yard is a potential source of diffuse emissions of tritium. This area is dedicated to hazardous-waste-, radioactive-waste-, and mixed-waste-management activities. The yard consists of several areas where waste containers are stacked outdoors. Many of these containers are not air tight and outgas tritium. A surveillance air monitor has been

placed in the Building 612 Yard to provide continuous measurements of tritium near this source. The median annual concentration of tritium in air for 1998 in this area was 130 pCi/m^3 (4.8 Bq/m^3). These data were used to calculate the total tritium emissions from the area, using a conservative approach that assumed the source to be 60 m south-southwest of the air sampler. With this assumption, a diffuse source emission of 4.6 Ci/y (1.75×10^{11} Bq/y) was required to produce the concentrations measured at the air sampler. This source term produced a calculated 1998 dose to the SW-MEI from the Building 612 Yard of 1.96×10^{-2} mrem (1.9×10^{-1} μSv).

Waste Accumulation Area Drum Sampling: Waste Accumulations Areas (WAAs) are maintained by the LLNL programs as storage areas for waste prior to the transfer of the waste to Hazardous Waste Management. Before the wastes are transferred, the waste drums are sampled by Hazardous Waste Management. Because this sampling represents a potential for exposure to the atmosphere, estimates of the potential dose from this activity are provided. The waste areas are maintained at various locations around the LLNL Livermore Site, so the potential emissions were modeled from the center of the site. This source term produced a calculated 1998 dose to the SW-MEI from the Waste Accumulation Areas (listed under Diffuse Sources, B612 in Attachment 1) of 5.1×10^{-4} mrem (5.1×10^{-3} μSv).

Southeast Quadrant: The Southeast Quadrant of the Livermore site has elevated levels of ^{239}Pu in the surface soil (from historic waste-management operations) and air (presumably from resuspension). A high-volume air-particulate sampler is located adjacent to the UNCLE Credit Union (the location of the SW-MEI) to monitor the ^{239}Pu levels in this area. Monitoring data from this air sampler were used as a direct measurement of potential dose via the air pathway. The median annual concentration of ^{239}Pu in air of 3.2×10^{-20} μCi/mL (1.2×10^{-15} Bq/mL), the dose-conversion factor of 3.08×10^5 mrem/μCi (8.33×10^{-5} Sv/Bq) from Federal Guidance Report No. 11, EPA-520/1-88-020, U.S. Environmental Protection Agency (1988) for ^{239}Pu, and the standard-man breathing rates of 8400 m^3/y were used to calculate the estimated EDE of 8.4×10^{-5} mrem (8.4×10^{-4} μSv) for 1998.

Site 300 Diffuse Sources

Diffuse sources at Site 300 involve tritium and uranium. During remediation efforts at Site 300, LLNL completed a contaminant screening to identify potential routes of migration from soil to air and other environmental media of these radionuclides and other contaminants (Final Site Wide Remedial

Investigation Report; Webster-Scholten, Ed., 1994, Lawrence Livermore National Laboratory, Livermore, CA, UCRL-AR-108131). Tritium and ^{238}U were identified as contaminants of potential concern.

Tritium Evaporation and Migration at Site 300: Tritium gas and solid tritium (Li^3H) were components of explosives assemblies tested on the firing tables during past experiments. Most of the gaseous tritium escaped to the atmosphere during the tests, but some of the solid Li^3H remained as residue in the firing table gravel. Rainwater and dust-control rinse water percolated through the gravel, causing the tritium to migrate into the subsurface soil and, in some cases, eventually to the ground water. Tritium-contaminated gravel was removed from the firing tables in 1988 and disposed in the Pit 7 landfill. Tritium in landfills, firing-table soils, and ground water are source terms for diffuse emissions of tritium to the atmosphere at Site 300. The tritium contamination at these locations was characterized at Site 300 in 1994. Since that time, natural processes including rainfall and evapotranspiration acted on the locations characterized, but new data have not been collected. Because it is becoming less likely that the 1994 data are representative of current conditions, LLNL personnel installed an air tritium sampler at a location (designated PRIM) that represents the SW-MEI, and doses from diffuse tritium sources for 1998 are estimated based on the monitoring data for that sampling location. The median annual concentration of tritium in air of 0.065 pCi/m^3 (2.4×10^{-3} Bq/m^3), the dose-conversion factor of 6.4×10^{-8} mrem/pCi (1.73×10^{-11} Sv/Bq) from Federal Guidance Report No. 11, EPA-520/1-88-020, U.S. Environmental Protection Agency (1988) for tritium, and the standard-man breathing rates of 8400 m^3/y were used to calculate the estimated EDE of 3.5×10^{-5} mrem (3.5×10^{-4} μSv) for 1998.

Resuspension of Depleted Uranium at Site 300: Like tritium, depleted uranium has been used as a component of explosives-test assemblies. It remains as a residue in surface soils, especially near the firing tables. Because surface soil is subject to resuspension by the action of wind, rain, and other environmental disturbances, the collective effects of surface soil uranium residuals on off-site doses were evaluated.

For the 1995 NESHAPs annual report, we developed calculations to separate the contribution to measured uranium activities from naturally occurring uranium (NU) (Gallegos et al., 1996, Lawrence Livermore National Laboratory, UCRL-ID-113867-96). We base our dose estimate for resuspended depleted uranium (DU) on the measured environmental surveillance

monitoring total concentration in air of uranium-238, subtracting out the part contributed by NU, from the following equation:

$$\mu = \frac{0.00726 - 0.99274\dfrac{M(CU-235)}{M(CU-238)}}{0.00526\dfrac{M(CU-235)}{M(CU-238)} + 0.00526} \ ,$$

where μ is the fraction (by weight) of uranium contributed by operations, CU is composite uranium (both DU and NU), M(CU-235) the mass of U-235 in the composite (measured) uranium, and M(CU-238) the mass of U-238 in the composite (measured) uranium. (For derivation of the equation see the 1995 NESHAPs annual report, referenced above.) This equation is used for those months in which explosives shots were not conducted.

Using these calculations to apportion the M(CU) for 1998, and excluding the appropriate months, we obtain an annual average concentration of DU in air from resuspension of 1.5×10^{-11} g/m^3. Using the fractions 0.998, 0.002, and 0.000005 to represent the amounts of ^{238}U, ^{235}U, and ^{234}U; specific activities of 3.32×10^{-7}, 2.13×10^{-6}, and 6.16×10^{-3} Ci/g for ^{238}U, ^{235}U, and ^{234}U; a yearly inhalation rate of 8400 m^3/y, and dose conversion factors from EPA Regulatory Guide 11 of 1.18×10^{11}, 1.23×10^{11}, and 1.32×10^{11} mrem/Ci; we obtain a total dose for resuspended DU of 5.3×10^{-3} mrem (5.3×10^{-2} μSv) for 1998.

Errata in 1997 Annual Report

In October 1998, LLNL revised its estimate of the radiological dose to the public from Site 300 operations for demonstration of compliance for calendar year 1997. The explanation and revisions were discussed in the letter of notification to EPA Region IX and are included here in Attachment 3.

Total Dose Estimate and Comparison with Previous Years' Data

For the Livermore site, the dose calculated for the SW-MEI from diffuse emissions in 1998 totaled 0.024 mrem (0.24 µSv). The dose due to point sources was 0.031 mrem (0.31 µSv). When combined, the total annual dose was 0.055 mrem (0.55 µSv). The relative contributions to the total were 43% from diffuse sources and 57% from point source emissions. The dose from point sources includes HT emissions modeled as HTO as directed by EPA Region IX. EPA Region IX acknowledges that such modeling results in a very conservative overestimation of the dose. This modeling methodology is used for purposes of compliance. However, we believe that this methodology does not provide a technically valid dose. A more accurate dose from both point and diffuse source emissions from the Livermore site is 0.049 mrem (0.49 µSv). Relative contributions to this total dose were 48% from diffuse sources and 52% from point source emissions.

The total dose to the Site 300 SW-MEI from operations in 1998 was 0.024 mrem (0.24 µSv). Point-source emissions from firing-table explosives experiments accounted for 0.019 mrem (0.19 µSv), or 78%, of this total, while 0.0053 mrem (0.053 µSv), or 22%, was contributed by diffuse sources. Table 5 presents the facilities or sources that account for 90% or more of the doses for the Livermore site or Site 300 SW-MEI.

Table 5. List of facilities or sources whose emissions account for 90% or more of the doses for the Livermore site and Site 300 SW-MEI.

Facility or Source	Dose (mrem)	Percent Contribution to Total Dose
Livermore site		
Building 331 (point source)	0.029 [a]	53%
Building 612 Yard (diffuse source)	0.019	35%
Building 331 Area Source (diffuse source)	0.0039	7%
Site 300		
Building 801 Firing Table (point source)	0.017	70%
Uranium resuspension (diffuse source)	0.0053	22%

a The dose includes HT emissions modeled as HTO as directed by EPA Region IX. EPA Region IX acknowledges that such modeling results in a very conservative overestimation of the dose. This methodology is used for purposes of compliance and we do not believe that it provides a technically valid dose estimate. The dose not having HT emission modeled as HTO is 0.023 mrem.

Comparison of the 1998 total dose estimate with that of previous years can be made by reviewing the information presented in Table 6. No diffuse emissions were reported at Site 300 for years before 1993, so comparison for total dose can only be made with the values for 1993 and later; in addition, diffuse source doses were not reported separately from the total dose for the Livermore site for 1990 and 1991.

Table 6. Doses (in mrem) calculated for the Site-Wide Maximally Exposed Individual for the Livermore site and Site 300, 1990 to 1998.

Year	Total Dose	Point Source Dose	Diffuse Source Dose
Livermore site			
1998	0.055 [a]	0.031 [a]	0.024
1997	0.097	0.078	0.019
1996	0.093	0.048	0.045
1995	0.041	0.019	0.022
1994	0.065	0.042	0.023
1993	0.066	0.040	0.026
1992	0.079	0.069	0.010
1991	0.234	—[b]	—[b]
1990	0.240	—[b]	—[b]
Site 300			
1998	0.024	0.019	0.005
1997	0.020	0.011	0.0088
1996	0.033	0.033	0.00045
1995	0.023	0.020	0.003
1994	0.081	0.049	0.032
1993	0.037	0.011	0.026
1992	0.021	0.021	—[c]
1991	0.044	0.044	—[c]
1990	0.057	0.057	—[c]

[a] The dose includes HT emissions modeled as HTO as directed by EPA Region IX. EPA Region IX acknowledges that such modeling results in a very conservative overestimation of the dose. This methodology is used for purposes of compliance and we do not believe that it provides a technically valid dose estimate. The total dose not having HT emission modeled as HTO is 0.049 mrem; the point source dose is 0.025 mrem.

[b] Diffuse source doses were not reported separately from the total dose for the Livermore site for 1990 and 1991.

[c] No diffuse emissions were reported at Site 300 for years before 1993.

SECTION V. Certification

I certify under penalty of law that I have personally examined and am familiar with the information submitted herein, and based on my inquiry of those individuals immediately responsible for obtaining the information, I believe that the submitted information is true, accurate, and complete. I am aware that there are significant penalties for submitting false information, including the possibility of fine and imprisonment.

Name: Phillip Hill
Deputy Director, Livermore Operations Division
U.S. Department of Energy
Livermore Site Office
7000 East Avenue, L-293
Livermore, CA 94550

Signature: _____ **Date:** _____
Phillip Hill

I certify under penalty of law that this document and all attachments were prepared under my direction or supervision in accordance with a system designed to assure that qualified personnel properly gather and evaluate the information submitted. Based on my inquiry of the person or persons who manage the system, or those persons directly responsible for gathering the information, the information submitted is, to the best of my knowledge and belief, true, accurate, and complete. I am aware that there are significant penalties for submitting false information, including the possibility of fine and imprisonment for knowing violations.

Name: L. Lynn Cleland
Livermore Site Manager
Laboratory Site Operations
Lawrence Livermore National Laboratory
7000 East Avenue, L-668
Livermore, CA 94550

Signature: _____ **Date:** _____
L. Lynn Cleland

SECTION VI. Supplemental Information

Collective Effective Dose Equivalent

Population doses, or collective EDEs, for both LLNL sites were calculated out to a distance of 80 km in all directions from the site-centers using CAP88-PC. As noted earlier, CAP88-PC evaluates the four principal exposure pathways: ingestion through food and water consumption, inhalation, air immersion, and irradiation by contaminated ground surface.

In 1996, we reconstructed the population distributions centered on the two LLNL sites. These population distributions, as were the previous distributions, are based on 1990 census data. However, the 1996 distributions were developed using commercially available, computer-map-based population data and the geographic information system software, ArcView©. The population for each sector segment was determined by selecting census block level data for that segment. In 1997, we further improved the estimates of the 1990 census population distribution by refining the location of the centers of the grids and by using curved arcs rather than straight lines to define the sector segments. Key population centers affected by LLNL emissions are the relatively nearby communities of Livermore and Tracy, and the more distant metropolitan areas of Oakland, San Francisco, and San Jose, as well as the San Joaquin Valley communities of Modesto and Stockton. Within the 80 km outer distance specified by the EPA, there are 6.3 million residents included for the Livermore site collective dose determination, and 5.4 million for Site 300. Our improved population data files (distribution of population with distance and direction) are shown in Tables 7 and 8 for the Livermore site and Site 300, respectively.

For the evaluation of the population dose, as distinct from the individual dose, all food (and in particular milk) was assumed to be produced locally. This decision was made because, although there are no commercial dairy animals within the distances used to evaluate individual doses, many dairy animals live within 80 km of the Livermore site and Site 300.

The collective EDE, which is the sum of the individual doses to all 6.3 million people within 80 km of the Livermore site, due to 1998 Livermore-site operations was 0.84 person-rem (0.0084 person-Sv). The collective dose includes HT emissions modeled as HTO as directed by EPA Region IX. EPA Region IX acknowledges that such modeling results in a very conservative overestimation of the dose. This methodology is used for purposes of

Table 7. Population distribution for LLNL's Livermore site, based on 1990 census information. Values are population in sector segments bounded by the indicated inner and outer radii, for each of sixteen 22.5°-sector directions.

	Range of distance from site (km)					
Direction	0-16	16-32	32-48	48-64	64-80	**Total**
N	235	12558	25414	6068	1932	**46207**
NNW	2135	1785	121044	1396	166741	**293101**
NW	6975	17085	247376	117130	102863	**491429**
WNW	1774	71710	224893	482899	152988	**934264**
W	49338	78214	312603	410117	568185	**1418457**
WSW	28590	115085	133563	311837	19824	**608899**
SW	304	85476	251417	129576	5113	**471886**
SSW	53	20234	600957	335772	59236	**1016252**
S	89	155	48296	61359	58915	**168814**
SSE	175	209	3	33	2481	**2901**
SE	321	55	50	25	9811	**10262**
ESE	139	166	1918	14064	55714	**72001**
E	77	7961	7103	153249	138118	**306508**
ENE	127	32766	60254	10831	3349	**107327**
NE	75	681	101717	219898	13442	**335813**
NNE	5	7115	1421	5570	18971	**33082**
Total	**90412**	**451255**	**2138029**	**2259824**	**1377683**	**6317203**

Table 8. Population distribution for LLNL's Site 300, based on 1990 census information. Values are population in sector segments bounded by the indicated inner and outer radii, for each of sixteen 22.5°-sector directions.

	Range of distance from site (km)					
Direction	0-16	16-32	32-48	48-64	64-80	**Total**
N	866	3363	2494	3633	6034	**16390**
NNW	104	4774	72306	4130	33751	**115065**
NW	88	225	25796	267551	107081	**400741**
WNW	152	20378	94428	309007	588389	**1012354**
W	454	72602	168776	285461	492124	**1019417**
WSW	49	43	188555	283552	123768	**595967**
SW	54	72	381738	641040	26040	**1048944**
SSW	4	3	46491	150412	24369	**221279**
S	19	242	3	26045	41175	**67484**
SSE	0	2	2	14	88	**106**
SE	33	15	151	8173	4938	**13310**
ESE	131	1286	13423	50535	32525	**97900**
E	270	2137	129980	133301	10026	**275714**
ENE	1264	21973	30017	22099	2845	**78198**
NE	32442	15122	87148	7502	4079	**146293**
NNE	4411	928	186995	69583	21515	**283432**
Total	**40341**	**143165**	**1428303**	**2262038**	**1518747**	**5392594**

compliance and we do not believe it provides a technically accurate dose estimate. The collective EDE not having HT emission modeled as HTO is 0.68 person-rem (0.0068 person-Sv). The collective dose is less than the 1997 value of 1.5 person-rem (0.015 person-Sv) because the stack releases from Building 331 (the Tritium Facility) decreased in 1998. This collective EDE can also be compared to the collective dose from natural background radioactivity for 6.3 million people of 1.88×10^6 person-rem (1.88×10^4 person-Sv).

The corresponding collective EDE from Site 300 operations in 1998, 11 person-rem (0.11 person-Sv), was due to point-source emissions. The total collective EDE value for Site 300 is higher than the 1997 value of 3.6 person-rem (0.036 person-Sv). The fact that this increase is not in direct proportion to the increase in the EDE at the SW-MEI is the result of differences in the amounts of high explosives and depleted uranium used each year in the individual explosives experiments.

The larger collective dose for Site 300 compared to the Livermore site is traceable primarily to the highly conservative assumptions about the Site 300 explosives experiments, especially regarding the fraction of radioactive material that is aerosolized and the height and trajectory of the explosive-debris cloud. This conservative modeling methodology over-predicts the quantity of radionuclides released to air by at least a factor of five, we believe, and over-estimates the long-range dispersal of material in these experiments. In 1992, we submitted to EPA a modeling protocol designed to treat the transient explosive experiments more realistically than does CAP88-PC, but this protocol was not accepted.

Compliance with 40 CFR 61 Subpart H (61.93)

Calculations of effective dose equivalents for all Livermore-site and Site 300 facilities having the potential to release radionuclides to the atmosphere have been completed. Annual doses from actual total emissions of all facilities during 1998 were found to be well below the 10 mrem (100 µSv) NESHAPs dose standard. Tritium accounted for most of the Livermore-site calculated dose, while at Site 300 practically the entire calculated dose was due to the isotopes ^{238}U, ^{235}U, and ^{234}U, in depleted uranium.

Based on potential emissions without control devices and EPA agreement, 17 emission points in three facilities at the Livermore site will maintain continuous monitoring systems in compliance with NESHAPs requirements. Continuous monitoring will be maintained in Building 332 and the seismically

hardened area of Building 251 instead of a modeling or measurement effort to demonstrate the actual need for monitoring. Continuous monitoring is being continued at Building 331 even though the EDEs that result from measured emissions do not require monitoring under 40 CFR 61.93(b).

Several other Livermore-site facilities (Buildings 175, 251 unhardened, 490, and 491) also will maintain continuous-monitoring systems; however, calculations using unabated potential emissions resulted in EDEs of less than 0.1 mrem/y (1 μSv/y) for the emissions from each of these facilities. While this monitoring also will be continued, it is not required under NESHAPs.

For facilities having discharge points without continuous monitoring, the requirement for continuous monitoring was individually evaluated. The evaluation was based on unabated emissions, even if emission-control systems existed. As a result of evaluations, two new continuous sampling systems, one at Building 292 and one at Building 177, were added in 1998. Building 292 involved a new operation and is discussed in Section IV "Construction and Modifications." At Building 177, a continuous filter-type sampler and multi-nozzle extraction probe was installed downstream of the HEPA-filtered exhaust from the Extractor Test Facility (ETF) to monitor for particulate emissions. Although ETF operations are not new, continuous sampling of the effluent was required because of plans for an increased usage inventory. Approval to construct or modify was not required because credit is allowed for the emission-control systems in that part of the regulation (40 CFR 61.96). No additional facilities at either LLNL site were found to require continuous monitoring.

Status of compliance with 40 CFR 61 Subpart Q - National Emission Standards for Radon Emissions from Department of Energy Facilities

LLNL does not have storage and disposal facilities for radium-containing materials that would be a significant source of radon.

Status of compliance with 40 CFR 61 Subpart T - National Emission Standards for Radon Emissions from the Disposal of Uranium Mill Tailings

LLNL does not have or store any uranium mill tailings.

Information on Radon-220 and Radon-222 Emissions

Radon emissions occur naturally by emanation from the earth. Radon-222 emissions that were reported in past NESHAPs annual reports from research experiments at the Livermore site did not occur in 1998.

1998 Air Monitoring

In this section we describe air effluent continuous sampling systems at LLNL facilities, periodic confirmatory measurements made in 1998 of emissions from sources not required to have continuous monitoring, and surveillance monitoring.

Continuous Monitoring: In 1998, there were eight buildings (Buildings 175, 177, 251, 292, 331, 332, 490, and 491) at the LLNL site that had radionuclide air

Table 9. Air-effluent sampling locations and systems.

Building	Facility	Analytes	Sample type	Number of samplers	Number of discharge points
175	MARS	Gross α, β on particles	Filter	6	6
177	ETF	Gross α, β on particles	Filter	1	1
251	Heavy Elements				
	Unhardened area	Gross α, β on particles	Filters	44	55[a]
	Hardened area	Gross α, β on particles	Filters	4	4
	Hardened area	Gross α, β on particles	CAM[b]	4	
292	Molten Salt Oxidation	Gross α, β on particles	Filter	1	1
331	Tritium	Tritium	Ionization Chamber[b]	4	2
		Gaseous tritium / tritiated water vapor	Molecular sieves	4	
332	Plutonium	Gross α, β on particles	CAM[b]	12	11
		Gross α, β on particles	Filters	16	
490	USEC Laser Isotope Separation	Gross α, β on particles	Filters	4	4
491	USEC Laser Isotope Separation	Gross α, β on particles	Filters	1	1

Note: "CAM" denotes Eberline continuous air monitors.
[a] Alternate blower system measured by the same sampler.
[b] Alarmed systems.

effluent monitoring systems. These buildings are listed in Table 9, along with the number of samplers, the types of samplers, the analytes of interest, and the number of monitored discharge points at the building. Many would operate from emergency power systems if normal power were lost.

Air samples for particulate emissions are extracted downstream of HEPA filters and prior to the discharge point to the atmosphere. Particles are collected on membrane filters. The sample filters are removed and analyzed for gross alpha and beta activity on a weekly or bi-weekly frequency depending on the facility. In most cases, simple filter-type aerosol collection systems are used. However, in some facilities, alpha continuous-air monitors (CAMs) are used for sampling. In addition to collecting a sample of particles, the CAM units provide an alarm capability for the facility in the event of a release of alpha activity.

Detection of gross alpha and beta activity resulting from particles collected on the air filters is accomplished using gas-flow-proportional counters. Analysis is delayed for at least four days from the end of sample collection to allow for the decay of naturally occurring radon daughters. For verification of the operation of the counting system, calibration sources, as well as background samples, are intermixed with the sample filters for analysis. Analysis is performed by the Radiological Measurements Laboratory (RML) in the Hazards Control Department (HCD).

Each stack of the Tritium Facility (Building 331) is monitored for tritium release by both a continuous-monitoring alarm system and continuous molecular-sieve samplers. The alarmed samplers, Overhoff ion chambers, provide real-time tritium concentration release levels (HT and HTO). The sieve samplers, which can discriminate between tritiated-water (HTO) vapor and molecular tritium (HT), provide the values used for environmental reporting and are exchanged weekly. Each sieve sampler (unalarmed) is in parallel with an alarmed monitor and consists of two molecular sieves. The first sieve collects tritiated water vapor; then a palladium-coated catalyst converts molecular tritium to tritiated water, which is then collected on a second sieve. The molecular sieve samples are submitted to the Hazards Control Analytical Laboratory where they are installed into a recovery system for the bake-out of tritiated-water vapor and subsequent condensation and collection of the water. The retrieved tritiated water is analyzed by RML using liquid-scintillation counting techniques.

Data from air-particulate-sampling filter and molecular-sieve analyses are reviewed by the Hazards Control Department Health Physicist responsible for

each facility and an Environmental Protection Department Environmental Analyst.

Periodic Confirmatory Sampling: Results of NESHAPs periodic confirmatory sampling serve to confirm two objectives: 1) that operations which are not continuously monitored do not need to be continuously monitored, and 2) that inventory-based estimates of emissions and their corresponding doses are conservative. In 1998, sampling was performed at certain operations in Buildings 177, 513, 625, and 854. The operations and sampling results are discussed below. None of the estimated emissions from any of the operations contribute significantly to the dose for the Livermore site SW-MEI. None of the operations require continuous sampling.

Building 177: At Building 177, we performed periodic confirmatory measurements for a vacuum pump exhaust from an operation using uranium. Three samples were taken, each on the pump down cycle of the operation. Samples of particulate emissions were collected on glass fiber filters. The filters were analyzed for gross alpha and gross beta activity. The average measured concentration was not distinguishable, or statistically different, from control measurements. (As for continuously monitored stacks, for which measured emissions are not significantly higher than background, these emissions are considered to be zero.) Since background concentration measurements have a lower limit of sensitivity that would result in a MEI dose far less than the 0.1 mrem requirement for continuous sampling, the exhaust from this operation does not require continuous sampling. Similarly, there is no significant contribution to the SW-MEI dose.

Building 513: We continued to make follow up air sampling measurements as a result of an unplanned release which occurred in the Building 513 Shredder Facility in July, 1997 (Gallegos et al., LLNL NESHAPs 1997 Annual Report, Livermore, CA, UCRL-ID-113867-98). In 1998, successive entries were made into the building and the shredder room to assess the contamination level of the room, to decontaminate the facility, and to remove the shredder. The nature of the entries made varied based upon the assessment plan of LLNL's Environmental Safety & Health Team of Hazards Control Division (HCD). As a result of the effort, the shredder and associated contaminated equipment were removed, and the radiological source term has been significantly reduced.

In support of the decontamination and decommissioning activities, which began in April 1998, and continued throughout the remainder of the year, weekly air samples were taken with a continuous air monitoring (CAM) unit located inside the shredder room and with a filter-type sampler located in an

adjacent room, the “cold” side. Gross alpha and gross beta activities were determined by analysis of the 47 mm diameter, AW-19 Millipore filters.

In addition to the room air concentration measurements, surveillance air sampling was conducted weekly outside the facility on the north side using high-volume air samplers at 3 locations for all of 1998. Three additional high-volume air samplers were positioned on the south side of the facility, but sampled only through mid-year. The north side locations were continued as the best monitoring points due to building access and prevailing wind direction. The surveillance samples were analyzed for ^{244}Cm using isotopic-specific chemical procedures and alpha spectroscopy.

Gross alpha and beta results from the room samplers indicated elevated air activity concentrations. The elevated levels could be directly correlated to shredder room activities conducted during the re-entry campaigns. Although the duration of room entries was limited to hourly intervals, sample collection was done on a weekly basis. Gross alpha activity was assumed to be dominated by ^{244}Cm. However, it is possible that a portion of the activity could be attributed to depleted uranium repackaging conducted prior to the incident. Gross beta activity was assumed to be attributed to a combination of depleted uranium and low-level isotopes contained within the HEPA filters which were being shredded at the time of the incident.

Modeling to estimate the public dose impact based on CAM sampling results was accomplished by calculating an annual source term from the average air activity concentration. The weekly room gross alpha activity concentrations from the CAM ranged from 6.0×10^{-4} pCi/m^3 to 1.3×10^{-1} pCi/m^3 with an average of 1.7×10^{-2} pCi/m^3. Using the stack ventilation rate and average air activity concentration, the annual source term was estimated to be 2.0×10^{-9} Ci. For purposes of CAP88-PC modeling, the alpha and beta activities were assumed to be ^{244}Cm and ^{90}Sr respectively. Results indicated an EDE of 1.6×10^{-6} mrem at the SWMEI and an EDE of 2.5×10^{-4} mrem (unabated) at the MEI.

Of the 121 environmental surveillance samples collected, seven yielded ^{244}Cm concentrations above detection limits. For these seven samples, concentrations ranged from 3.1×10^{-6} pCi/m^3 to 2.6×10^{-5} pCi/m^3 with an average of 1.2×10^{-5} pCi/m^3. The shredder room concentrations ranged 200 to 5000 times higher that those observed by the environmental surveillance monitors for the minimum and maximum concentrations, respectively. It can be speculated that fugitive “hot” particles leaking from the building during decontamination entries account for the samples that had detectable values.

Air surveillance sampler results were also compared to the air activity concentrations modeled using the same source term described above. The modeled air activity concentrations of ^{244}Cm ranged from 1.6×10^{-6} pCi/m^3 to 1.6×10^{-5} pCi/m^3 and compared favorably with the air surveillance sampler results. In this case, an area source type was assumed in CAP88-PC because the samplers were located close to the source (within 20 m) and concentrations derived from CAP88-PC using the plume source type are suspect within 25 m from the source. Using the area source methodology resulted in an EDE of 1.9×10^{-6} mrem at the SW-MEI and 3.5×10^{-3} mrem (unabated) at the MEI.

Both approaches yielded similar annual doses to the SW-MEI. Also, neither approach indicates that continuous air sampling of the operation is required according to NESHAPs. For reporting purposes, we have been conservative by choosing the higher SW-MEI estimate, or 1.9×10^{-6} mrem. This EDE value is less than 1% of the estimated EDE reported for the shredder incident in 1997.

Building 625: At Building 612, we sampled the HEPA-filtered exhaust from decontamination activities being conducted in a tent-like enclosure. Sampling took place for 1 week as decontamination activities were taking place. Air samples of the exhaust were extracted isokinetically and collected on Millipore 47-mm diameter, AW-19 filters. The filters were analyzed for gross alpha and gross beta activity. The average measured air concentrations were 9.2×10^{-23} Ci/mL and 1.1×10^{-22} Ci/mL for gross alpha and beta activity respectively. Using the ventilation rate from the process and assuming the activities continued the entire year, gross alpha and gross beta emissions were estimated to be 1.4×10^{-9} Ci and 1.8×10^{-9} Ci respectively. By CAP88-PC modeling the resulting dose to the SW-MEI would be 3.7×10^{-6} mrem. The dose to the MEI, 4.0×10^{-6}, is far less than 0.1 mrem so that continuous monitoring of the operations is not required. The estimated dose to the SW-MEI based upon the usage inventory of the operations was 4.3×10^{-13} mrem. Although this dose was less than the dose based on the periodic measurements, it must be realized that both dose estimates are near zero.

Building 854: At Site 300, historical shot samples containing mixed fission products and actinides packaged in sealed drums were opened for sampling purposes. Operations were conducted in a glove box within a tent enclosure. A continuous air monitor (CAM) was used to sample the HEPA-filtered exhaust from the operation. The 47-mm diameter, membrane filters were analyzed for gross alpha and gross beta activity. The operation was of short duration, lasting only a week. The average measured air activity

concentrations were 7.3 x 10^{-20} Ci/mL and1.2 x 10^{-19} Ci/mL for gross alpha and beta activity respectively. Using the process ventilation rate, estimated emissions for the one-week operation were 2.1 x 10^{-8} Ci gross alpha activity and 3.5 x 10^{-8} Ci gross beta activity. Modeling the gross alpha activity as ^{239}Pu and the gross beta activity as ^{90}Sr, the resulting EDE estimates were 1.0 x 10^{-6} mrem at the MEI location and 4.7 x 10^{-7} mrem at the SW-MEI location. These EDE estimates were much less than inventory-based evaluations which resulted in 2.9 x 10^{-2} mrem and 4.0 x 10^{-2} mrem at the MEI and SW-MEI locations respectively. Thus, the inventory approach was conservative.

General Surveillance Monitoring: Surveillance air monitoring for tritium and radioactive particles has been in place since the 1970s and will continue. LLNL currently maintains eight continuously operating, high-volume, air-particulate samplers on the Livermore site, nine in the Livermore Valley, eight at Site 300, one offsite near Site 300, and one in Tracy. LLNL also maintains eleven continuously operating airborne-tritium samplers on the Livermore site, six samplers in the Livermore Valley and one offsite near Site 300. The samplers are positioned to ensure reasonable probability that any significant airborne concentration of particulate and tritium effluents resulting from LLNL operations will be detected. Many of the surveillance air monitors are placed near diffuse emission sources, such as those near Buildings 292, 331, 513, 514, and 612, as well as in and around the Southeast Quadrant of the Livermore site. As such, their results can be used to estimate and/or confirm the emissions from the associated diffuse sources. Also included are air-particulate and air tritium monitors positioned at the locations of the SW-MEI for the Livermore site and Site 300. Results from the latter samplers provide a source term for large area diffuse sources and also serve to confirm the SW-MEI EDEs as determined from facility emissions using air effluent monitoring results and usage inventories.

The data from the air surveillance monitoring network provide continuous measurements of the concentrations of radionuclides present in the air at the Livermore site, Site 300, and in the surrounding areas. Data from the network are presented in the LLNL Environmental Report, which is prepared annually and available to the public. (Larson et al., Environmental Report for 1998, Lawrence Livermore National Laboratory, Livermore, CA, UCRL-50027-98, to be published in October 1999.)

Comparison of 1998 Modeling Results with Surveillance Monitoring Data

A comparison was made between CAP88-PC modeling results and surveillance air monitoring data for all eleven tritium air monitors on the Livermore site (designated VIS, SALV, POOL, CAFE, MESQ, MET, COW, B331, B514, B624, and B292) and one off-site tritium air monitor (ZON7). Monitor locations are shown in Figure 6.

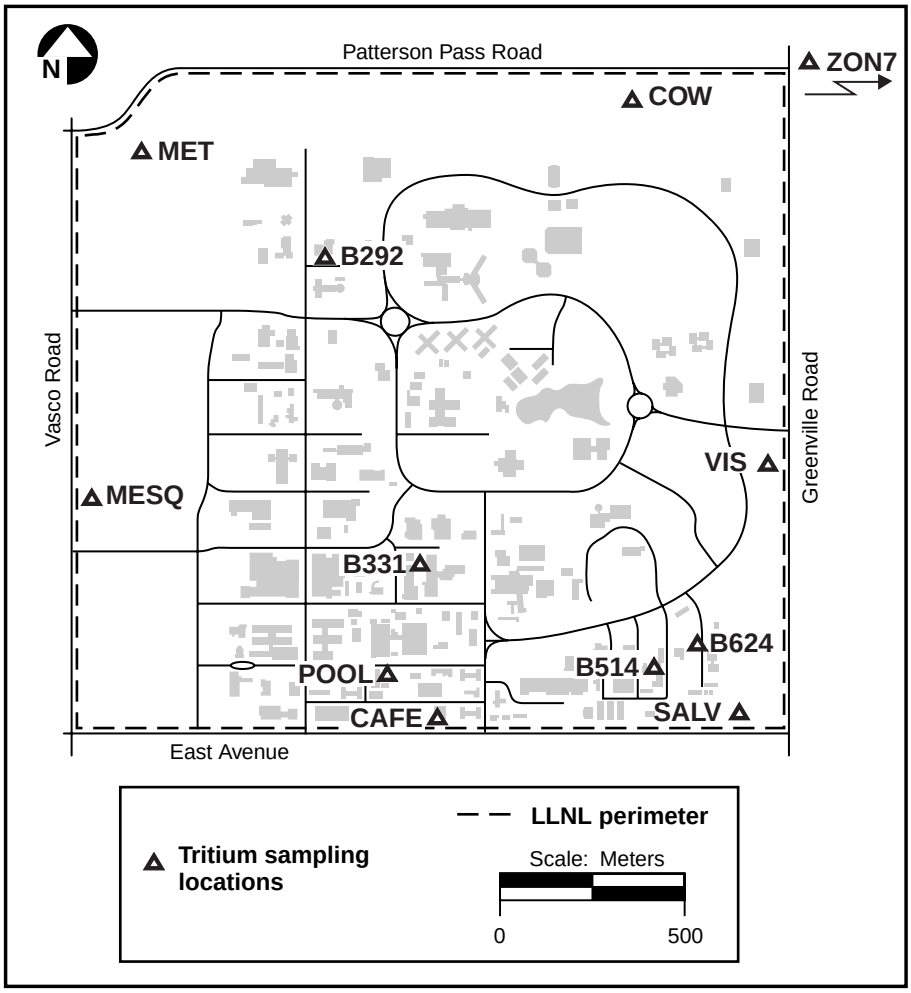

Figure 6. Tritium air-surveillance sampling locations.

Only the three most significant sources of tritium releases to air at the Livermore site were included in the model-data comparison. The largest source is the Tritium Facility (Building 331), where the tritium is emitted from two 30-m-high, continuously monitored stacks; a total of 85 Ci (3.1 TBq) of HTO was emitted from these stacks in 1998. The other two principal

sources are diffuse areas associated with the Building 612 yard and Tritium Facility (Building 331) yard. Emissions from these sources were estimated to be 4.6 Ci (0.17 TBq) and 6 Ci (0.22 Tbq) in 1998. All other potential sources of tritium release, such as the hazardous waste management operations in Building 514 and the Building 292 diffuse source were too minor to influence the model-data comparison.

Annual-average concentrations (number of pCi/m^3 of air) at the locations of the twelve monitors were calculated for the three sources individually and collectively, and compared to the measured annual median concentrations at the twelve monitoring locations. The results are displayed in Figure 7.

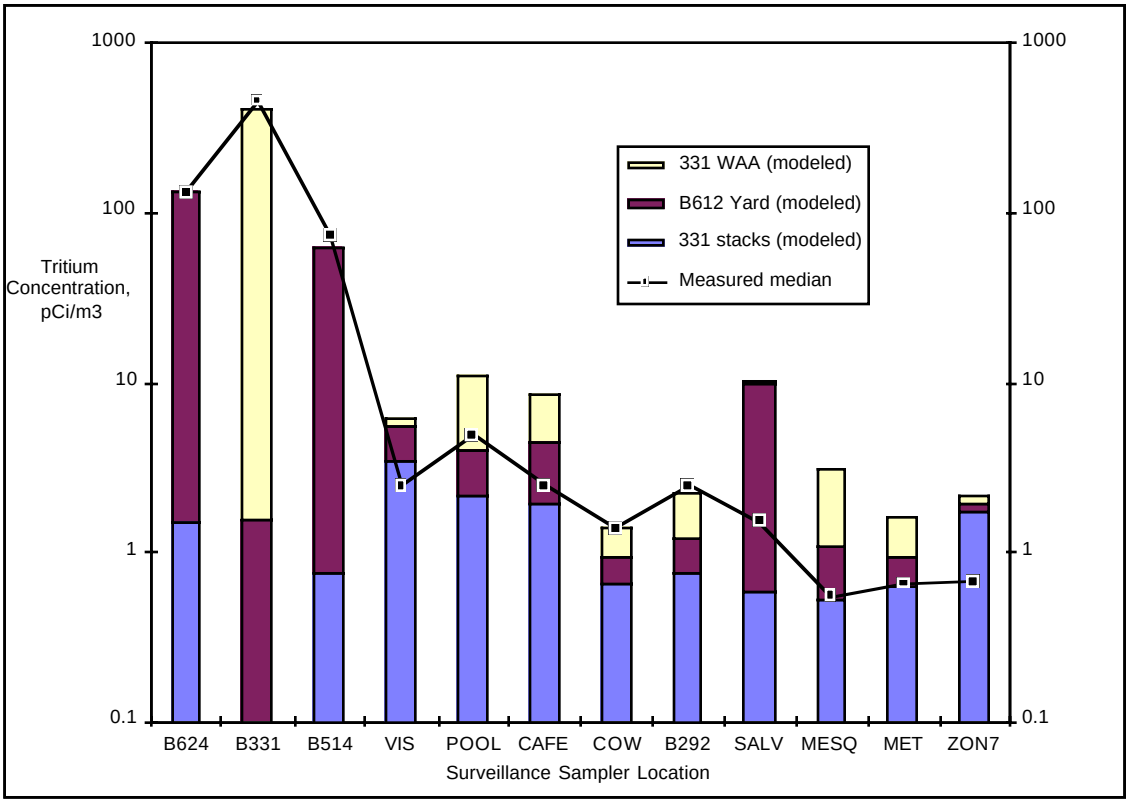

Figure 7. Comparison of measured and modeled tritium concentrations, 1998. Note that the logarithmic scaling used visually distorts the smaller concentration values.

The Building 331 stack emissions were used as input to CAP88-PC with the site-specific meteorological data to calculate the annual-average

concentrations at the desired locations. However, both the B331 Yard and the B612 Yard emission rates were not independently measured, but rather were determined from the surveillance tritium air monitor data for the particular monitor in the closest proximity, by requiring that the modeled concentration match the data from that particular monitor. The source term for Building 612 Yard was adjusted to give the observed value at the B624 monitor, and the source term for the B331 Yard was chosen to give agreement with the measured value at the B331 monitor; in each case the diffuse source was the dominant source for the monitor in question, accounting for more than 95% of the measured concentration in the nearby monitor. Using this approach, the modeling results, by design, agree with the monitoring data at the B624 and B331 locations.

The main conclusion shown in Figure 7 is that by taking into account the three leading sources of tritium releases to air—the Building 331 stacks, Building 612 Yard, and the Building 331 Yard—fairly good agreement is obtained with data for all of the monitors. Generally, the modeling results agree with the on-site monitoring data within a factor of 3.5 (at 10 out of twelve locations). However, in the case of two on-site monitors (SALV and MESQ), the difference is nearly a factor of seven, with the model resulting in higher concentration predictions.

Status of the NESHAPs QA Program

The LLNL NESHAPs Quality Assurance (QA) Program is a multi-organizational effort that relies on the Quality Assurance/Quality Control programs that are in place at the LLNL facilities with continuous air-monitoring systems, the Radiological Measurements Laboratory (RML) and the Analytical Laboratory of the Hazards Control Department (HCD), and the Environmental Protection Department (EPD). Memoranda of understanding (MOUs) are in place between EPD and the facilities and/or programs and HCD; these MOUs formalize responsibilities and obligations of the organizations regarding many tasks for the air effluent sample network. Tasks that are addressed include air sampler design and installation, procedures and their implementation, sampling, sample analysis and tracking, maintenance and repair of sampling systems, guidance on regulatory requirements, documentation of the sampling network, reporting, and the archival of records.

Facility Safety Procedures (FSPs), Safety Analysis Reports (SARs) and QA Manuals for monitored facilities describe their organizational structures, responsibilities for sampling locations used for continuous air monitoring,

and the procedures to be followed in the case of unplanned radionuclide releases. For example, the FSP for the Plutonium Facility (Building 332) describes in detail the procedure for responding to detection of radioactive materials in a release from the stacks. These documents also describe the sample-collection systems for both real-time and passive (i.e., not alarmed) air-monitoring systems, and procedures to be used for measuring flow rates, sampling, and calibration.

The RML Quality Assurance Program describes laboratory-analysis procedures, precision, accuracy and completeness objectives, sample-tracking procedures, quality-control (QC) sampling, sample handling, and data reporting. For example, the Gross Alpha-Beta Procedures Manual of the RML describes operational procedures for analyzing the air sampler filters for radioactivity.

EPD, which is responsible for NESHAPs modeling and reporting, also operates under a Quality Assurance Management Plan and associated procedures. Detailed records are kept of all measurements, CAP88-PC model runs, and calculations, and selected model runs are validated. The Terrestrial and Atmospheric Monitoring and Modeling Group (TAMM) of EPD is responsible for modeling and reporting radionuclide emissions for NESHAPs compliance. TAMM members continue to refine mechanisms that ensure they are informed whenever new operations are proposed, significant changes in radionuclide inventories occur, or existing operations are modified so that NESHAPs modeling can be performed and appropriate action taken. All NESHAPs calculations are archived with the supporting information used to make the calculations.

LLNL has drafted a quality assurance project plan (QAPP) which assembles the quality assurance methods including the above information into one complete document. The document is structured similarly to that specified for a quality assurance program contained in Appendix B, Method 114 of 40 CFR 61. It describes the organization structure and functional responsibilities, objectives of the quality assurance program, administrative controls in place for handling unplanned emissions, sample collection, analysis and tracking procedures, sample collection systems, sample collection and effluent flow rate measurement systems, corrective actions, and reporting. The draft QAPP is currently under review by EPD.

Quality Control (QC) for 1998 Radiological Inventory Update and Modeling

Radiological Inventory Update QC: Approximately 15% of the 62 potential discharge points that completed radiological-inventory updates in 1998 were randomly selected for validation. For this QC check, radiological inventories from nine potential emission points were selected for validation: two from Building 132N; one from Building 151; one from Building 222: three from Building 177; one from Building 514; and one from Building 612. An EPD Environmental Analyst contacted the responsible party who signed the NESHAPs Inventory Forms and physically visited and inspected the facilities to verify inventory data. The responsible party was asked to demonstrate how he/she arrived at the data submitted on the original inventory form. Stack parameters also were verified. The QC data were compared to the original data. The accuracy of the inventory data was confirmed.

Modeling QC: Fifteen percent of the CAP88-PC modeling runs were selected for validation by a second analyst using a different computer and copy of CAP88-PC. The analyst performing this QC effort ran the model following independent gathering of radionuclide inventories and stack data from the NESHAPs Inventory Forms and pertinent distances from site maps. The QC modeling verified the values from the original CAP88-PC modeling runs. The data that are presented in the attached spreadsheet are as accurate as possible, demonstrating that quality objectives are being met.

EPA Compliance Evaluation Investigation

There were no compliance evaluations of LLNL facilities in 1998.

Attachment 1. 1998 LLNL NESHAPs Annual Report Spreadsheet

Guidance for Interpreting Attachment 1

A generalized description of each facility and its operations is provided on the spreadsheet. In addition, the following information is shown for each listed emission point or stack:

- Building and room number(s)
- Specific stack identification code(s)
- Generalized operations in the room(s) or area(s)
- Radionuclides utilized during 1998
- Annual radionuclide inventory with potential for release (by isotope, in curies)
- Physical-state factors (by isotope)
- Stack parameters
- Emission-control devices and emission-control-device abatement factors
- Estimated or measured annual emissions (by isotope)
- Distance and direction to the site-wide maximally exposed individual (SW-MEI)
- Calculated EDE to the SW-MEI
- Distance and direction to the maximally exposed individual for that specific source (MEI)
- Calculated EDE to the MEI (source term not adjusted for emission controls)
- Source category

Radionuclides: The radionuclides shown in the spreadsheet are those from specific emission points where air emissions were possible. If radionuclides were present, but encapsulated or sealed for the entire year, radionuclides, annual inventories, and emissions are not listed.

Radionuclide Inventories with Potential for Release: The annual radionuclide inventories for point-source locations are based on data from facility experimenters and managers. For Buildings 251 (hardened area) and 332, classification issues regarding transuranic-radionuclide inventories make use of the inventory/modeling approach impractical. However, all such affected emission points in these buildings are continuously monitored, and emissions are therefore directly determined. LLNL conducted a complete radionuclide-inventory update in 1997.

Physical-State Factors: The physical-state factors listed are EPA potential-release fractions from 40 CFR 61, Appendix D, whereby emissions are estimated from radionuclide inventories depending on their physical states for use in dispersion/dose assessment modeling. A physical-state factor of 1.0×10^{-6} is used for solids, 1.0×10^{-3} is used for liquids and powders, and 1.0 is used for unconfined gases. In 1996, U.S. EPA granted approved alternative emissions factors for elemental uranium as follows: an emission factor of 1×10^{-6} can be used for elemental uranium heated at temperatures below 1100°C, an emission factor of 1×10^{-3} can be used for elemental uranium heated at temperatures below 3000°C, and an emission factor of 1 shall be used for temperatures greater than 3000°C. These factors are allowed provided that the uranium is not intentionally dispersed to the environment and that the processes do not alter the chemical form of the uranium.

Stack Parameters: Engineering surveys conducted from 1990 through 1992 form the basis for the stack physical parameters shown, which were checked and validated by facility experimenters and managers for 1994 and 1995. Stack physical parameters for sources evaluated in 1998 were updated, as necessary, by experimenters and managers for those facilities.

Emission-Control Devices: High-Efficiency-Particulate-Air (HEPA) filters are used in many LLNL facilities to control particulate emissions. For some discharge points, scrubbers and electrostatic precipitators aid the control of emissions. The operational performance of all HEPA filtration systems is routinely tested. The required efficiency of a single-stage HEPA filter is 99.97%. Double-staged filter systems are in place on some discharge points. Triple-stage HEPA filters are used on glove-box ventilation systems in the Building 332 Plutonium Facility and in the hardened portion of Building 251.

Control-Device Abatement Factors: Similar to physical-state factors, control-device abatement factors, from Table 1 in 40 CFR 61, Appendix D, are those associated with the listed emission-control devices, and are used to better estimate actual emissions for use in dispersion and dose models. By regulation, each HEPA filter stage is given a 0.01 factor (even though the required test efficiency that all LLNL HEPA filters must maintain would yield a factor of 0.0003).

Estimated Annual Emissions: For unmonitored and non-continuously monitored sources, estimated annual emissions for each radionuclide are based on the product of (1) inventory data, (2) time factors (discussed in "Emission Source Terms" of in Section III, (3) EPA potential-release fractions

(physical-state factors), and (4) applicable emission-control-device abatement factors.

Actual emission measurements are the basis for reported emissions from continuously monitored facilities. LLNL facilities that have continuous monitoring systems are Buildings 175, 177, 251, 292, 331, 332, 490, and 491. See the subsection titled "1998 Inventory Update and Effective Dose Equivalent (EDE) Calculations" for a discussion of the use of emissions measurements for monitored sources.

10 mrem/y Site-Wide Dose Requirement: For LLNL to comply with the NESHAPs regulations, the LLNL site-wide maximally exposed individual (SW-MEI; defined as the hypothetical member of the public at a single residence, school, business, or office who receives the greatest LLNL-induced EDE from the combination of all radionuclide source emissions) cannot receive an EDE greater than 10 mrem/y (100 μSv/y).

In Attachment 1, the distance and direction to the respective SW-MEI are shown for each facility at each site. Doses to the site-specific SW-MEIs were evaluated for each source and then totaled for site-specific evaluations against the 10 mrem/y dose standard (see "Total Dose Estimate" in Section IV).

0.1 mrem/y Monitoring Requirement: To assess compliance with the requirement for continuous monitoring (potential dose greater than 0.1 mrem/y [1.0 μSv/y]), emissions must be individually evaluated from each point source; the location of the maximally exposed public individual (MEI) is generally different for each emission point. The maximum dose at a location of unrestricted public access typically occurs at a point on the site perimeter. Therefore, it is often referred to as the maximum "fence line" dose, although the off-site maximum dose could occur some distance beyond the perimeter. (This could happen, e.g., when the perimeter is close to a stack; however, for all emission points at the Livermore site and Site 300, calculations show that ground-level concentrations of radionuclides decline monotonically beyond LLNL boundaries.) As stipulated by the regulations, modeling for assessment of continuous monitoring requirements assumed unabated emissions (i.e., no credit was taken for emission abatement devices, such as filters), but physical-state factors were applied.

The unabated EDE cannot be calculated for monitored facilities. Because the monitoring equipment is placed after HEPA filtration, there is no way to obtain an estimate for what the emissions might have been had there been no filtration. It is not reasonable to apply factors for the effects of the HEPA filters

on the emission rate because most of what is measured on the HEPA filters is the result of the radioactive decay of radon, which is capable of penetrating the filter. Attachment 1 gives, for each inventoried point source, the dose to the MEI and the distance and direction to the LLNL fence line where the MEI is located. However, for monitored sources, no value is shown.

Source Categories: LLNL radionuclide air-emission sources have been classified into seven source categories, indicated by the number in the next to last column of the spreadsheet: (1) Unmonitored or non-continuously monitored Livermore-site facilities that have had a radionuclide-inventory update for 1998; (2) Unmonitored or non-continuously monitored Livermore site facilities with a previous radionuclide-inventory update (this category is not used in years with complete inventory updates, like 1997); (3) Continuously monitored Livermore-site facilities; (4) Site 300 explosives experiments; (5) Diffuse sources where emissions and subsequent doses were estimated using inventory processes; (6) Diffuse sources where emission and dose estimates were supported by environmental-surveillance measurements; and (7) Sources whose emissions estimates and subsequent doses were estimated based on periodic confirmatory air sampling rather than continuous sampling.

(this page intentionally left blank)

Attachment 2. Surrogate Radionuclides List

Although CAP88-PC supports calculations for many radionuclides, there are some in use at LLNL that are not included in CAP88-PC. This list of surrogate radionuclides has been developed to account for the contribution of those radionuclides. In addition, isotopic analyses of mixtures of radionuclides are not always available, and radionuclide inventories are stated as "gross alpha," "gross beta," "gross gamma," or "mixed fission products" (MFP). In these cases, ^{239}Pu was used as the surrogate for gross alpha, ^{137}Cs was used as the surrogate for gross gamma, and ^{90}Sr was used as the surrogate for gross beta and mixed fission products to provide conservative dose estimates.

Table 2-1. List of surrogate radionuclides.

Isotope	Half-Life	Lung Class	ALI (inh) µCi	DAC (inh) µCi/cm^3	Surrogate	Half-Life	Lung Class	ALI (inh) µCi	DAC (inh) µCi/cm^3
Ag-108m	127 y	Y	2.0×10^{1}	1.0×10^{-8}	**Co-60**	5.271 y	Y	3.0×10^{1}	1.0×10^{-8}
Bi-207	38 d	W	4.0×10^{2}	1.0×10^{-7}	**Bi-214**	19.9 min	W	9.0×10^{2}	4.0×10^{-7}
Ca-45	163 d	W	8.0×10^{2}	4.0×10^{-7}	**Sr-90**	29.12 y	D	2.0×10^{1}	8.0×10^{-9}
Cd-109	464 d	Y	1.0×10^{2}	5.0×10^{-8}	**Co-60**	5.271 y	Y	3.0×10^{1}	1.0×10^{-8}
Cf-249	350.6 y	Y	1.0×10^{-2}	4.0×10^{-12}	**Cm-245**	8500 y	W	6.0×10^{-3}	3.0×10^{-12}
Cf-250	13.1 y	W	9.0×10^{-3}	4.0×10^{-12}	**Am-241**	432.2 y	W	6.0×10^{-3}	3.0×10^{-12}
Cl-36	3.01×10^{5} y	W	2.0×10^{2}	1.0×10^{-7}	**Cs-137**	30 y	D	2.0×10^{2}	6.0×10^{-8}
Es-254	275.7 d	W	7.0×10^{-2}	3.0×10^{-11}	**Pu-239**	24065 y	Y	2.0×10^{-2}	7.0×10^{-12}
Eu-149	93.1 d	W	3.0×10^{3}	1.0×10^{-6}	**Pm-151**	28.4 hr	Y	3.0×10^{3}	1.0×10^{-6}
Gd-148	93 y	D	8.0×10^{-3}	3.0×10^{-12}	**La-140**	40.272 h	W	1.0×10^{3}	5.0×10^{-7}
Os-185	94 d	D	5.0×10^{2}	2.0×10^{-7}	**Mo-99**	66 h	Y	1.0×10^{3}	6.0×10^{-7}
P-33	25.4 d	W	3.0×10^{3}	1.0×10^{-6}	**P-32**	14.29 d	D	9.0×10^{2}	4.0×10^{-7}
Re-184	38 d	W	1.0×10^{3}	6.0×10^{-7}	**Mo-99**	66 h	Y	1.0×10^{3}	6.0×10^{-7}
Se-75	119.8 d	W	6.0×10^{2}	3.0×10^{-7}	**As-76**	26.32 h	W	1.0×10^{3}	6.0×10^{-7}
Sr-85	64.8 d	D	3.0×10^{3}	1.0×10^{-6}	**Sr-90**	29.12 y	D	2.0×10^{1}	8.0×10^{-9}
Ta-182	115 d	Y	1.0×10^{2}	6.0×10^{-8}	**Hf-181**	42.4 d	W	4.0×10^{2}	2.0×10^{-7}
Tb-157	110 y	W	3.0×10^{2}	1.0×10^{-7}	**La-140**	40.272 h	W	1.0×10^{3}	5.0×10^{-7}
Tb-158	180 y	W	2.0×10^{1}	8.0×10^{-9}	**La-140**	40.272 h	W	1.0×10^{3}	5.0×10^{-7}
Tl-204	3.78 y	D	2.0×10^{3}	9.0×10^{-7}	**Pb-214**	26.8 min	D	8.0×10^{2}	3.0×10^{-7}
Tm-168	93.1 d	W	2.0×10^{3}	8.0×10^{-7}	**La-140**	40.272 h	W	1.0×10^{3}	5.0×10^{-7}
Tm-171	1.92 y	Y	3.0×10^{2}	1.0×10^{-7}	**La-140**	40.272 h	W	1.0×10^{3}	5.0×10^{-7}
Y-88	106.64 d	Y	2.0×10^{2}	1.0×10^{-7}	**Y-90**	64 h	Y	6.0×10^{2}	3.0×10^{-7}
Am-244	10.1 h	W	2.0×10^{2}	8.0×10^{-8}	**Cm-244**	18.11 y	W	1.0×10^{-2}	5.0×10^{-12}
Au-195	183 d	Y	4.0×10^{2}	2.0×10^{-7}	**Ba-133**	10.74 y	D	7.0×10^{2}	3.0×10^{-7}

(continued ...)

Table 2-1. List of surrogate radionuclides (concluded).

Isotope	Half-Life	Lung Class	ALI (inh) µCi	DAC (inh) µCi/cm³	Surrogate	Half-Life	Lung Class	ALI (inh) µCi	DAC (inh) µCi/cm³
Co-56	78.76 d	Y	2.0×10^{2}	8.0×10^{-8}	**Co-60**	5.271 y	Y	3.0×10^{1}	1.0×10^{-8}
Gd-146	48.3 d	W	3.0×10^{2}	1.0×10^{-7}	**Sm-147**	1.06×10^{11} y	W	4.0×10^{-2}	2.0×10^{-11}
Kr-85	10.72 y	Gas	See Note	1.0×10^{-4}					
Rh-102	2.9 y	Y	6.0×10^{1}	2.0×10^{-8}	**Rh-106m**	29.9 s	Y	4.0×10^{4}	1.0×10^{-5}
U-239	23.54 min	Y	2.0×10^{5}	6.0×10^{-5}	**U-240**	14.1 h	Y	2.0×10^{3}	1.0×10^{-6}
Zr-90	809 ms	W	N/A	N/A	**Y-90**	64 h	Y	6.0×10^{2}	3.0×10^{-7}

Note: The DAC for Kr-85 also has been relaxed considerably since its beta emission only skin. The DAC is based on limitation of non-stochastic effects in the skin; the MPC w assuming that the beta particles of energy greater than 0.1 MeV contributed to the whc

[a] D = days, W = weeks, Y = years.

Source: Limiting Values of Radionuclide Intake and Air Concentration and Dose Conversion Factors for Inhalation, Submersion and Ingestion, Federal Guidance Report No. 11, EPA-520/1-88-020, U.S. Environmental Protection Agency, 1988.

Attachment 3. Revised Estimate of Dose for 1997 Site 300 Operations

OAKLAND OPERATIONS OFFICE

University of California
Lawrence Livermore
National Laboratory

Laboratory Site Operations
October 23, 1998

David Howekamp
Director, Air Division
U.S. Environmental Protection Agency
75 Hawthorne Street
San Francisco, CA 94105

Dear Mr. Howekamp:

Lawrence Livermore National Laboratory (LLNL) has revised its estimate of the radiological dose to the public from Site 300 operations for demonstration of compliance with NESHAPs for calendar year 1997. The revision of the dose was prompted by the identification of a miscommunication of location data used in assessing the radiological dose from firing table operations. LLNL recently notified EPA Region IX of the results of the dose reassessment by telephone and email. This letter confirms those communications and provides additional detail.

Unfortunately, the identification of this issue occurred after June 1998, when the final LLNL NESHAPs report was submitted to your office. Attachment A is a summary of the corrections to the LLNL NESHAPs 1997 Annual Report as a result of the reassessment. The significant changes are as follows:
1) the dose from firing table operations increased to 0.011 mrem, from 0.0054 mrem,
2) the total dose to the site-wide maximally exposed individual (MEI) from all Site 300 operations increased to 0.02 mrem, from 0.014 mrem, and
3) the population dose increased to 7.2 person-rem, from 3.6 person-rem.

The dose to the site-wide MEI from Site 300 operations remains small compared to the annual standard of 10 mrem per year.

Last minute efforts by LLNL were successful in incorporating these changes in LLNL's *Environmental Report 1997*, released to the public on October 1, 1998. In the near future, this letter and its attachment will be added to the electronic version of LLNL NESHAPs 1997 Annual Report available over the Internet, as well as sent to all recipients of the 1997 annual report on our distribution list. Finally, the information will also appear as an errata section in the 1998 NESHAPs annual report.

L. Lynn Cleland
Laboratory Site Manager,
Lawrence Livermore National Laboratory

Phillip E. Hill
Director,
Livermore Operations Division
Department of Energy

Attachment

TAMM98_037/AB/LLC/PEH/jk

Attachment 3.

cc:	Ballard, E.	DOE/LSO, L-293
	Corey, R.	DOE/LSO. L-293
	Fisher, D.	L-005
	Galles, H.	L-626
	Jackson, S.	L-633
	Lasell, S.	DOE/OAK
	Lee, J.	L-701
	Surano, K.	L-629
	Sutherland, D.	DOE/LSO, L-293
	DCC	

TAMM98_037/AB/LLC/PEH/jk

Attachment A
Corrections to 1997 NESHAPs Annual Report (UCRL-ID-113876-98)

The following corrections were necessitated by recalculation of emissions from experimental tests at Site 300.

1. Page 1, Second bullet
Corrected text
Site 300: 0.020 mrem (0.20 μSv) (55% from point-source emissions, 45% from diffuse-source emissions)
Previous text
Site 300: 0.014 mrem (0.14 μSv) (38% from point-source emissions, 62% from diffuse-source emissions)

2. Page 8, Second paragraph
Corrected text
Similarly, detailed information is given in Attachment 1 for experiments at a Site 300 explosives-testing facility (Building 801 and its associated firing tables).
Previous text
Similarly, detailed information is given in Attachment 1 for experiments at two Site 300 explosives-testing facilities (Buildings 850 and 851 and their associated firing tables).

3. Page 17, Fourth paragraph
Corrected text
The calculated EDE to the SW-MEI at Site 300 was calculated to be 0.011 mrem (0.11 μSv) from point-source emissions. All of this EDE resulted from Building 801 firing-table emissions in the course of explosives experiment; there were no explosives experiments at Building 851 in calendar year 1997. The 1997 EDE is a decrease from the 0.033 mrem (0.33 μSv) dose modeled for 1996. The decrease in dose is primarily the result of a decrease in the number of experiments that contained depleted uranium in 1997.
Previous text
The calculated EDE to the SW-MEI at Site 300 was calculated to be 0.0054 mrem (0.054 μSv) from point-source emissions. All of this EDE resulted from Building 850 and Building 851 firing-table emissions in the course of explosives experiments—52% from the former and 48% from the latter. The 1997 EDE is a decrease from the 0.033 mrem (0.33 μSv) dose modeled for 1996. Building 801, which accounted for more than one-half of the dose in 1996, was not in use in 1997 due to construction activities at the Contained Firing Facility being built at that location (discussed below). The decrease in dose is

primarily the result of a decrease in the number of experiments that contained depleted uranium and, to a lesser extent, the result of the fact that Buildings 850 and 851 are farther from the fence line than Buiding (*sic*) 801.

4. Page 28, Third paragraph, fourth and fifth sentences

Corrected text

The total dose to the Site 300 SW-MEI from Site 300 operations in 1997 was 0.020 mrem (0.20 µSv). Point-source emissions from firing-table explosives experiments accounted for 0.011 mrem (0.11 µSv), or 55%, of this total while 0.0088 mrem (0.088 µSv), or 45%, was contributed by diffuse sources.

Previous text

The total dose to the Site 300 SW-MEI from Site 300 operations in 1997 was 0.014 mrem (0.14 µSv). Point-source emissions from firing-table explosives experiments accounted for 0.0054 mrem (0.054 µSv), or 38%, of this total while 0.0088 mrem (0.088 µSv), or 62%, was contributed by diffuse sources.

5. Page 29, Table 5, Site 300 data

Corrected text

Site 300		
Uranium resuspension (diffuse source)	0.0087	44%
801 Firing Table (point source)	0.011	55%

Previous text

Site 300		
Uranium resuspension (diffuse source)	0.0087	61%
850 Firing Table (point source)	0.0028	20%
851 Firing Table (point source)	0.0026	18%

6. Page 29, Table 6, Site 300 1997 data

Corrected text

Site 300			
1997	0.020	0.011	0.0088

Previous text

Site 300			
1997	0.014	0.0054	0.0088

7. Page 33, Second paragraph, first sentence

Corrected text

The corresponding collective EDE from Site 300 operations in 1997, 7.2 person-rem (0.072 person-Sv), was due to point-source emissions.

Previous text

The corresponding collective EDE from Site 300 operations in 1997, 3.6 person-rem (0.036 person-Sv), was due to point-source emissions.

8. Page 61, Site 300 Point Source Section, Building 850 and 851 Firing Tables

The corrections for this information is presented differently, due to the large format of the table on that page.
The information for Buildings 850 and 851 should be deleted.
The following information for Building 801 should be added, for the headings that are listed in **bold type**.
Building 801; **Room/Area** Firing Table; **Operation** Explosive tests; **Radionuclides** U-238, U-235, U-234; **Annual Inventory with Potential for Release (Ci)** 5.4e-02, 7.0e-04, 5.0e-3 (for U-238, U-235, U-234, respectively); **Physical State Factor** 1.0e00, 1.0e00, 1.0e00 (for U-238, U-235, U-234, respectively) **Stack Height (m)** NA; **Stack Diameter (m)** NA; **Stack Velocity (m/s)** NA; **Control Devices** None; **Estimated Annual Emissions** 5.4e-02, 7.0e-04, 5.0e-3 (for U-238, U-235, U-234, respectively); **Distance to SW-MEI (m)** 2380; **Direction to SW-MEI** ESE; **EDE (mrem)** 0.011; **Distance to MEI (m)** 1809; **Direction to MEI** ENE; **Unabated EDE (mrem)** 0.018; **Source Category** 4; **Below App. E Quantity** Y.